



Report on the Paducah Gaseous Diffusion Plant Metals Recovery Program



*Department of Energy
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ACRONYMS

AEC	Atomic Energy Commission
AMESQ	Assistant Manager for Environment Safety, Health and Quality
CEDE	Committed Effective Dose Equivalent
CIP/CUP	Cascade Improvement and Cascade Upgrade Program
DOE ORO	Department of Energy, Oak Ridge Operations
DRS	Decon and Recovery Services, Inc.
ERDA	Energy Research and Development Administration
ETTP	East Tennessee Technology Park
IBA	Invitation, Bid and Acceptance
ICP	Inductively Coupled Plasma
K-25	Oak Ridge Gaseous Diffusion Plant
KOW	Kentucky Ordinance Works
LMUS	Lockheed Martin Utility Services
MDA	Minimum Detectable Activities
MMUS	Martin Marietta Utility Services
NLO, Inc.	Department of Energy Fernald Plant
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PGDP or Plant	Paducah Gaseous Diffusion Plant
PGDP	Paducah Gaseous Diffusion Plant
U. S.	United States
UCC-ND	Union Carbide Corporation-Nuclear Division
Union Carbide	Carbide and Carbon Chemicals Company
USEC	United States Enrichment Corporation
USEC	United States Enrichment Corporation

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1.0 EXECUTIVE SUMMARY

e Metals were processed and recycled at PGDP from 1952 to 1986.

The Department of Energy, Oak Ridge Operations (DOE ORO), conducted an investigation of the Paducah Gaseous Diffusion Plant (PGDP or Plant) Metals Recycling Program from August 25, 1999 - October 31, 1999. The review was conducted to determine if contaminated metals were released into commerce. Metals processed from 1952 to 1986 included: steel, nickel, aluminum, copper, monel (a copper-nickel alloy), cobalt, and precious metals (gold and silver). Extensive reviews were made of historical documentation, interviews were held with current and retired employees, and analyses were conducted of the remaining or connected materials.

Based on available records, between 2,800 and 5,300 lbs. of gold were recovered and shipped from the PGDP between 1964 and 1985. The process used for reclaiming the gold was kept separate from other materials and processes that were possibly contaminated, but the process was conducted in contaminated areas of Buildings C-400 and C-746A. The worst-case use of gold would have been through pharmaceutical injection in arthritic patients. If this material were used for this purpose, it would have resulted in an exposure of about 30 mrem, or 10% of annual natural background. But this exposure scenario is extremely unlikely.

Approximately 7,650 lbs. of silver were reclaimed from the reprocessing of classified x-ray film from 1966 to 1974. The film was burned in the Building C-405 incinerator and the ash was smelted into silver bars in the Building C-



Exhibit 1 - Paducah Gaseous Diffusion Plant

727 Foundry.

The film and process would not have been contaminated unless residual contamination was picked up from contaminated facilities. This is corroborated by analysis of a composite sample of several spools of x-ray film discovered in the PGDP laboratory. However, cross-contamination could have occurred during processing. One slag sample analyzed showed technetium-99 (^{99}Tc) to exceed background; however, there is no evidence to indicate any potential existed for cross-contamination at a level that would possibly be of concern.

Large quantities of clean and contaminated nickel powder were recovered by smelting into ingots in Building C-746A. One or more sample buttons were made with each pour. Approximately 17 million lbs. of clean nickel were released into commerce. Nickel free of radioactivity and that which was below the AEC and site criteria were routinely

transferred to the National Stockpile or sold.

Nearly 20 million lbs. of contaminated nickel were cast. None of this material was sold or otherwise released from radiological control, and the material is currently maintained in the PGDP contaminated scrap yard.

Samples of “clean” ingots sold into commerce contained low levels of both ^{99}Tc and plutonium-239 (^{239}Pu) contamination. However, these levels would have had no public health consequence.

Approximately 4.5 million lbs. of aluminum were smelted into ingots in Building C-746A from 1970-1986. Records prior to 1984 could not be found. Analysis of aluminum samples has revealed low levels of thorium-230 (^{230}Th), ^{239}Pu , ^{240}Pu , and uranium-233 (^{233}U). The actinide levels present in the aluminum would not have resulted in health consequences to the general public. However, the potential to exceed annual radiation protection standards for the public might possibly have existed at foundries where this aluminum was remelted.

Scrap steel was segregated into “contaminated” or “clean” by process knowledge and spot-check surveys; however, no documentation about this is available. It is possible that contaminated steel could have been sold. Interviews with staff indicate that contaminated steel was sometimes found with clean steel. Excess clean steel was sold on the open market. If contaminated steel was identified, it was moved to the contaminated scrap yard and was not sold. Approximately 26.7 million lbs. of contaminated steel scrap were generated. A small portion was smelted, however, process problems caused the smelting operation to be terminated. The ingots produced remain with the contaminated steel scrap.

Excess copper was shipped to NLO, Inc. (the DOE Fernald Plant) for reprocessing. After

economic reevaluation the decision was made to establish copper processing at the East Tennessee Technology Park (ETTP) in Oak Ridge. The entire inventory was shipped to ETTP and is presently being processed and sold. The copper is surveyed prior to sale and a separate verification survey is carried out by a different contractor.

Small quantities of cobalt and monel have been smelted and sold. Although both metals were associated with the manufacture of new, uncontaminated barrier and other processes, one record shows that in 1986 monel was melted with contaminated nickel in a ratio of 55 to 1 and sold. No record exists on the contaminant level of the nickel; thus it is impossible to calculate the level in the monel.

Lead was recycled including weapons parts. The only information available concerned the sale of shredded lead. Records show that approximately 258,990 lbs. of shredded lead were produced and released into commerce. There would have been a slight potential for cross-contamination of the lead during its processing at PGDP.

In certain instances, this report raises uncertainties with respect to the impact of metals released to the public. The best available information is presented here so that management can determine where further investigation is warranted.

2.0 INTRODUCTION

The DOE ORO, conducted an investigation of the PGDP Metals Recycling Program from August 25, 1999 - October 31, 1999. The review was conducted to determine if contaminated metals were released into commerce as alleged (i.e., gold, silver, aluminum, nickel, monel, cobalt, copper, lead, steel). This report documents the findings from that review.

NOTE: Throughout the remainder of the report, references to C-400, C-746, etc., refer to buildings.

e The purpose of this investigation was to determine if contaminated metals were released into commerce.

2.1 INVESTIGATIVE APPROACH

To support the overall objective of determining whether contaminated metals were released off-site, historical documentation for the metal's program was requested from Bechtel Jacobs Company, LLC (BJC LLC), and the United States Enrichment Corporation (USEC). To facilitate these requests, BJC LLC and USEC established points of contact. A number of requested documents could not be located. These include:

- perpetual inventories of precious metals;
- copies of all procurement documents associated with sale of gold and silver: (1) sales agreements on gold, and (2) registers or logs for shipping and receiving during the 1950s, '60s, '70s, and '80s dealing with precious metals (the team found a small

sample of shipping orders involving gold; no silver bar procurement information was found or provided);

- backup files from *Committee Report on Weapons Scrap Program*; the Committee's findings were transmitted to A. M. Tuholsky on May 7, 1982;
- *Disposal of the PGDP Scrap Metal*, Report Number KY/F-127, PGDP Scrap Disposal Committee, dated April 1980;
- radiation and contamination surveys of gold recovery and smelting equipment between 1965 to the present (not including surveys done as requested by the team.);
- radiation surveys or radio chemical analyses (1960 to the present) used to determine radiological purity, or for free-release, of smelted metals (gold, nickel, aluminum, copper, lead, cobalt, silver, steel, monel) that were released for unrestricted use (the team found some radio chemical analyses for aluminum in their records search);
- all photographs related to the gold smelting process in C-400 during the 1970s and '80s (the team found some photographic negatives of the gold process in the C-100 vault);
- annual *Percent Gold Yield* reports;
- *Au (gold) Record Book*; and
- Standard Operating Procedure CH-359, *Silver Recovery in C-400* (the team was provided a copy through the Department of Justice).

An indexed evidence file of documents that were provided by BJC LLC, USEC, or otherwise found by the team has been maintained.

During the course of the investigation, it was discovered that files from the C-746A smelter had been placed in drums for disposal as low-level hazardous waste. Several hundred drums of

this waste were stored in C-746A. These drums had been used for clean-out of the facility several years ago. A search was made of the disposal records covering a window of time from before C-746A closed (December 1974) until several years after closure. The document search revealed 23 drums which contained items listed as “paper, trash, etc.” These drums were pulled and searched by the team in the C-746B warehouse. Six of the drums contained file records, operational logbooks, engineering drawings, and other information pertaining to the Metals Recycling Program, particularly information on nickel, silver, aluminum, and gold. Several color and black/white photographs were also found. Numerous documents were turned over to Bechtel Jacobs for health physics survey, classification review, and copying. Copies of the requested documents were later provided to the team. However, one document that the team had noted on cobalt was not included with the copied material and could not be found.

In an effort to capture as much historical information as possible, an e-mail was sent to all Bechtel Jacobs and USEC employees. This notice requested employees with knowledge about certain aspects of the Metals Recycling Program to contact the team (**Attachment 1.**)

Numerous interviews were conducted with USEC and Bechtel Jacobs employees, retired workers, representatives of the U. S. Department of the Treasury, Westpoint Mint in New York, and representatives of five companies identified as having purchased gold recovered at Paducah. A significant number of interviews were conducted with retired employees who had worked in the early metals recovery programs.

Wherever available, the team obtained metal samples, process residues, and equipment

surveys in order to assess the extent of any radioactive contamination currently present and the possibility that metals may have been cross-contaminated by past operations. Sample buttons obtained from nickel and aluminum melts were still stored onsite. The team requested independent analysis for a random, representative number (120) of the sample buttons from lots that had been sold. Of those buttons requested, Bechtel Jacobs found 80 samples. These were analyzed by gamma spectroscopy and/or inductively coupled plasma (ICP) mass spectrometry for radioisotopes including ^{99}Tc , uranium-238 (^{238}U), and neptunium-237 (^{237}Np). Neither historical radio chemical data nor samples were available for the precious metals. Therefore, surveys and analyses were conducted on available process residues (slag from melts, ash, dissolver crud) and leachates from equipment that contacted the metals during processing. Residue samples and/or leachates were also analyzed for radionuclides by ICP mass spectrometry. Results from the various analyses are given in **Attachment 2.**

Evidence collected from interviews, documentation, and sample analyses were used to document if the potential existed for PGDP to have added any radioactive contamination to recycled metals released into commerce.

2.2 HISTORY OF THE PGDP

The PGDP is located in McCracken County, Kentucky, approximately 10 miles west of the city of Paducah and 3 miles south of the Ohio River. The site occupies 3,425 acres, of which 750 acres are within a security fence and contains uranium enrichment process equipment and support facilities. The current mission of the Plant is to “enrich” uranium for use in domestic and foreign commercial power reactors.

Production of enriched uranium began in the early 1940's as a United States defense initiative to produce fissionable material for the atomic bomb. All of the enrichment facilities and the weapons plant at Oak Ridge, Tennessee, and the labs and production facilities in the West were initially included in this defense initiative. Later, the nuclear weapons program including the enrichment facilities were transferred to the Atomic Energy Commission (AEC). The Production Division within the Oak Ridge office of the AEC was responsible for all operational facilities including all of the enrichment facilities. The country's first gaseous diffusion plant, the Oak Ridge Gaseous Diffusion Plant (K-25), went on line in 1945.

On December 15, 1950, the National Security Resources Board chose the Paducah site for the second gaseous diffusion plant. The site chosen was the old Kentucky Ordinance Works (KOW). The KOW was a production facility for gunpowder and TNT for World War II and resided on more than 5000 acres. The KOW above-ground structures were dismantled and a new government facility was begun almost simultaneously. Early construction utilized some of the KOW infrastructure for both construction and early operation of the Paducah Plant.

e From 1952 until the late 1980s, different programs other than diffusion were conducted at the Plant.

Initial Plant construction required major support infrastructure including shops that had cutting-edge equipment to construct and maintain diffusion equipment that was larger and more intricate than anything available at the time. As the Plant went into operation cell by cell, the equipment in the two sets of cascade buildings required significant support from plant

maintenance and machine shop crews. However, the level of effort required to maintain the Plant was less than had been needed during construction. This created a manpower issue for each diffusion plant. Therefore, the very close ties with Oak Ridge and the many secure facilities connected with the war efforts of the past decade (1940s) led the Paducah Plant to begin an effort to assist with work at other facilities. This work included such things as fabricating parts for nuclear and non-nuclear weapons, dismantling of weapons, recycling of classified materials, and other technical services in specialized areas.

During those early days (1950s and 1960s), the Plant was secured and protected the same as any of the other facilities involved in the war effort. The remote location, the quality and availability of the workforce, and the relatively low cost of labor suited these efforts perfectly. Over the period of time from 1952 until the late 1980s, several different programs were conducted at the Paducah Plant that were separate from the diffusion mission. For example, Paducah was designated as the center for metals recycling and recovery by the AEC in 1967.

Federal presence began with the defense initiatives during the early 1950s and evolved with a significant AEC staff and military presence through the late 1950s. With the AEC, a significant staff continued into the 1960s. After the mid-1960s, the AEC staff dwindled until the last federal employee onsite retired in late 1978. From 1978 until 1987 there was no onsite federal staff at PGDP. All management was performed from Oak Ridge. The existing DOE Site Office began in January 1987 when the first permanent DOE employee arrived onsite to start and expand the office.

Carbide and Carbon Chemicals Company (now Union Carbide) was the original site contractor

and operated PGDP for the AEC. In the mid-1960's, the mission of the Plant shifted from military to commercial applications, and the Plant began enriching uranium for use in nuclear power plants. In 1975, the Energy Research and Development Administration (ERDA) was formed to assume responsibility for regulatory oversight of the uranium enrichment program. In 1977, ERDA became DOE. In 1984, Martin Marietta Energy Systems, Inc., became the operating contractor for PGDP, replacing Union Carbide Corporation-Nuclear Division (UCC-ND).

The Energy Policy Act of 1992 created the United States Enrichment Corporation (USEC) and was a first step in the process of privatizing the government's uranium enrichment enterprises. In July 1993, USEC leased portions of the Plant from DOE, assumed responsibility for uranium enrichment activities, and contracted with Martin Marietta Utility Services (MMUS), a subsidiary of Martin Marietta, for operation and maintenance of enrichment activities. Through a June 1995 corporate merger, MMUS became Lockheed Martin Utility Services (LMUS). The Nuclear Regulatory Commission (NRC) granted a certificate of compliance to the Plant in November 1996, under 10 CFR Part 76, and regulatory oversight of enrichment operations was transferred from DOE to the NRC in March 1997. As a result of an initial public offering, USEC was privatized as an investor-owned corporation in July 1998. USEC took over

direct operation of all enrichment activities at the Plant in May 1999, with most LMUS personnel becoming employees of USEC.

DOE retains responsibility for the environmental restoration program, most elements of the waste management program, and all waste materials

generated by past DOE (and predecessor) activities. BJC LLC, is the management and integrating contractor for DOE for the remainder of the Plant, having been awarded this contract in April 1998. Bechtel Jacobs relies on subcontractors to conduct environmental restoration and waste management functions. USEC facilities consist of process buildings, electrical switch yards, steam plant, water treatment facility, chemical cleaning and decontamination facility, and maintenance and laboratory facilities.

3.0 METALS RECYCLING PROGRAM

3.1 BACKGROUND

Construction, maintenance and refurbishment of U. S. uranium enrichment facilities have generated large quantities of scrap metals. These metals were supplemented by U. S. weapons program scrap beginning in 1964. The types of scrap metals were predominantly nickel, aluminum, copper, and steels. Classified scrap metals and film containing silver were smelted to destroy classified attributes. Scrap metals that met release criteria (current for that period) were recycled and released to the public.

In the 1970s, a Cascade Improvement and Cascade Upgrade Program (CIP/CUP) generated large amounts of scrap nickel, aluminum, steel, and copper (mostly electric cabling and motor windings). Disposition of scrap materials from the program activities was by recycle, disposal, or storage for future recycle. Nearly all of the scrap stored for future recycle is still located at DOE facilities.

Generic limits for volumetric contamination have never been established. Historically, however, AEC apparently had the authority to approve the release of volumetrically contaminated metals on a case-by-case basis. DOE 5400.5 currently allows for the derivation and approval of volumetric contamination limits on a case-by-case basis. None were ever solicited or approved for the PGDP recycled metals.

Information was collected to document the regulatory requirements and authority for the release of surface-contaminated scrap metal from AEC/ERDA/DOE sites for the period in question. The earliest correspondence cite the requirements and authority of AEC Manual Chapter 5170.

e Weapons parts were received for dismantling and disposal.

On July 13, 2000, the Secretary of Energy suspended all metals recycling activities within DOE following complaints from the public, the metals industry, and labor unions at DOE contractor sites.

3.2 GOLD

3.2.1 Material Processing Description

In the early days, weapons components were sent to PGDP for dismantling and disposal. After working on this task for several years, tests were conducted on reclaiming gold. The tests were successful and in 1964 Paducah began removing parts that contained gold from the incoming scrap. PGDP tracked scrap by supplier, which provided a "path" back to that supplier for the funds that remained after the cost for recovery was calculated and subtracted. The recovery of gold in most cases provided funding for the total cost of the weapons scrap dismantling and declassification.

The gold recovery process is outlined in PGDP procedure, CH-358, *Gold Recovery in C-400*. The steps outlined in this procedure were corroborated with interviews and document reviews. After accumulation of gold-bearing scrap in C-746A, the material was transported in lots to the C-400 gold recovery area located in the west bay at the north end of the building. When received, the Chemical Operations Department prepared a *Rare and Precious Metal Certificate* (Form UCN-1672). The material was then processed in a nitric acid solution in the Gold Scrap Dissolver Tank (**Exhibit 2**).



Exhibit 2 - Gold Scrap Dissolver Tank.

The tank had steam heating coils to increase the action of the nitric acid. The gold scrap remained in this tank for several hours and occasionally overnight to allow for full dissolution of the scrap. Everything except plastic and gold was dissolved. Gold leaf that floated on top of the solution was skimmed off and the nitric acid solution was decanted into the filtrate settling tank; the remaining solids were placed on the gold wash table (**Exhibit 3**). The table had a recirculating water system to continually flush water over the materials removed from the Gold Scrap Dissolver Tank. A screen at the deeper end of the wash table would accumulate gold scrap that was freed from the dissolved parts. This gold was placed in cheesecloth-like “socks,” dried in an oven, and compressed into a small cylinder



Exhibit 3 - Gold Wash Table.

“hamburger” by a hand press. This “hamburger” was then placed in a crucible for melting in a small furnace (**Exhibit 4**).

The gold accumulated from the gold wash table would be melted and poured into a small bar, usually between 20 and 50 ounces. A heavy screen was used when pouring the gold to catch any dross or slag. The slag left from the pouring of the bar of gold was accumulated and remelted to remove any remaining gold. The re-melt of slag may have occurred several times before the process removed all of the available gold. Any gold that could not be removed on the gold wash table and any remaining slag left after several re-melts were totally dissolved in an aqua regia solution. Because this process would dissolve everything, including gold, copper strips would be dissolved into the solution to aid in the precipitation of the gold. After precipitation, the gold recovered was then re-melted and poured into small bars. This part of the process produced gold of the highest purity (99.9%).

After several smaller bars of gold were produced, these bars would be re-melted into a larger bar. The objective was to ultimately

pour an approximate 300-ounce bar of gold



Exhibit 4 - Small furnace used to smelt reclaimed gold.

(Exhibit 5), which was preferred by the Treasury Department. These bars were then shipped directly to the New York Assay



Exhibit 5 - 300 oz bar of gold.

Office, U. S. Department of Treasury for refinement. The Assay Office acted as the AEC's agent in the sale of the refined gold until the late 1970s when the gold standard was removed. Union Carbide then, through some unclear association with the Treasury Department, began selling gold to commercial reproprocessors.

e The New York Assay Office acted as AEC's agent in the sale of the gold until the late 1970s.

3.2.2 Quantities of Material Recovered

- c Gold was recovered from weapons assemblies and scrap parts from July 1964 until October 1985.
- c Between 2,800 and 5,300 pounds of gold was recovered at PGDP.

The team was unable to determine the exact amount of gold produced at PGDP due to the unavailability of complete records. Two important

documents in determining the amount of gold processed were the Gold Processing Records and the Gold Bar Summary. The Gold Processing Records indicated that gold-plated scrap was received from five primary sources: (1) Medina-Clarksville, (2) Burlington Amarillo Military Sites, (3) DASA-Kelly, (4) DASA-Other, and (5) Burlington/Pantex. The record also indicates that approximately 306,578 lbs. of gold-plated scrap was received and processed. The total amount of gold recovered, according to the Gold Processing Records, totals 5,283.66 lbs. (77,299.92 troy ounces).

The Gold Bar Summary lists gold bars and their appropriate weights from number 1 to 102. The total gold produced according to this log is 1,739.37 lbs. (25,446.95 troy ounces). During extensive records search, the team discovered a number of handwritten sheets listing bar numbers and associated weights. These bar numbers started at number 71 and went through bar 159. A comparison of the sheets for bar numbers 71 through 102 on the Gold Bar Summary were identical. This confirmed to the team that more than 102 bars of gold were produced. However, the team could not confirm if more than 159 bars were produced. After the site copied and transferred the handwritten records to Oak Ridge for classification review, sheets for bar numbers 153 through 159 were missing. Additional search of records in the C-710, Records Vault, at the site did not produce the missing sheets. Using the information from the handwritten sheets (with an average weight for the missing bar sheets), the total gold produced in the 159 bars was approximately 2,817.54 lbs. The team concludes that the total gold produced is probably more accurately expressed in the Gold Processing Records since the total information contained in the records is considerably more detailed than in the Gold Bar Summary.

3.2.3 Disposition of Material

- C Recovered gold was sold by PGDP or sold through the New York Assay Office. Documentation indicates that some of the gold was transferred to the Y-12 Plant in Oak Ridge and some was used in the Paducah Machine Shop in C-720.

Upon completion of the gold recovery process and production of gold bars, the bars were sent to stores inventory for storage and safeguarding pending disposition. The team attempted to trace the gold through the entire procurement process and requested the site to produce all procurement records associated with these transactions. The team searched and located shipping orders and related procurement information for some of the gold transactions. Based on these few records, it appears that the procurement flow was as follows: UCC-ND (later MMES and LMES) prepared an Invitation, Bid and Acceptance (IBA) document for sale of government-owned property via the sealed bid process. Purchasing assigned P-000 numbers to each bid package as the controlling number. General sales terms and conditions were printed on the reverse of the IBA. A Special Provisions document was generated for each IBA. In addition to other procurement requirements, the document listed the location of the gold. Upon receipt of sealed bids, an award was made to the highest bidder by the Purchasing Department. A data sheet was prepared identifying the bidder name and relevant bid information.

Five companies were identified that purchased gold and are still in existence. Using a specific script (**Attachment 3**), contact was made with these companies: Waterbury Companies, Inc., Waterbury, Connecticut; Technic, Inc., Cranston, Rhode Island; Westbury Alloys, Inc., Westbury,

New Jersey; Handy and Harmon Refining, Attleboro, Massachusetts; and Leach and Garner Refining Corp., North Attleboro, Massachusetts. None of the companies had records of the Paducah gold purchase. However, each company reported that the routine procedure when gold is received is to perform a purity analysis on the gold. Additional refining is then performed and all of the gold in stock at the time is consolidated. The major markets for the gold are the jewelry and the banking industries. The companies reported that no radioactive analysis of gold was performed by them during the time-frame of the Paducah gold purchases.

Records indicate that at least three gold bars were transferred to the Oak Ridge Y-12 Plant in 1981. However, all attempts to track the gold were unsuccessful. Also, records indicate that gold was transferred to the Paducah Machine Shop in C-720. While no records could be located to determine its use, pictorial evidence indicates a potential weapons-related purpose.

e Incoming weapons components were not surveyed at PGDP until 1982.

3.2.4 Analytical Results

- C No documented evidence was found that gold was surveyed or analyzed for radioactive contamination prior to sale or release from the site.

An operations manager indicated that no laboratory analyses were conducted to detect the presence of radionuclides in the gold. He recollected that the gold may have been

surveyed using hand-held instruments, but the results were not documented. The use of the hand-held survey instruments was confirmed by

a previous health physics technician. This technician did not remember ever finding evidence of contamination using these instruments.

- C Incoming weapons components were not surveyed at PGDP until 1982.

In 1982, Pantex accidentally shipped some radioactive parts to PGDP that were mixed with normal non-radioactive weapons scrap. The radioactive parts were discovered when a manager surveyed the scrap out of curiosity. However, the radioactive material found in 1982 was not associated with the gold components, and there is no record of any incident involving the gold.

- C Crucibles used to melt the gold, and molds into which the gold was poured revealed the presence of depleted uranium and technetium-99 contamination.

Because actual samples from the gold bars are not available, items that came in direct contact with the molten gold or the gold bars were surveyed and analyzed. Leachates and residues from crucibles used in the gold furnace as well as molds into which the gold was poured were analyzed by mass spectrometry. Results from leachates from used crucibles and molds were compared with those of an unused crucible (control) that had been stored in the same location.

The leachate from the unused crucible contained 15 pCi of ^{238}U . All other isotopes in the leachate were below the detection limits for the analysis.

NOTE: For sample results from ICP mass

spectrometry, uranium results are given for ^{238}U alone. Since depleted uranium constituted the vast majority of material processed in the Feed Plant and Greensalt Plant (C-410/420) and/or fed to the PGDP cascade from 1952 through 1980, it is conservative to assume that the total uranium concentrations in this report are roughly double those for the reported ^{238}U values.

The two black crucibles that had been used in gold melts had leachate levels of 84 and 110 pCi ^{238}U , respectively. Comparing this maximum value with that of the control crucible leachate, it is possible that the uranium levels seen in the leachates resulted from crucible usage during gold production.

Solid residues from the black crucibles were obtained after leaching. These residues indicated much higher levels of contamination. ^{238}U levels in the residues ranged from 480 to 718 pCi/g, and ^{99}Tc levels ranged from 15,000 to 32,000 pCi/g. These levels exceed what one would expect from surface deposition of contamination alone while the crucibles were stored in C-400. Other isotopes in the residues were below the detection limits of the mass spectrometry analysis. The small masses of the residues (0.13 g and 0.25 g) severely limited the ability to detect ^{237}Np . On the larger sample, the ^{237}Np result was reported as less than 34 pCi/g. (In **Attachment 2.2**, both the residue leachate and digestion results from black crucible #69977 were each less than 280 pCi/g for ^{237}Np . More sensitive re-analysis of the archived residue samples showed the residue contained a less than 34 pCi/g total ^{237}Np activity concentration).

Leachates from the two gold molds revealed ^{238}U levels of 380 and 530 pCi, respectively.

Given that the molds had been stored for many years inside of a contamination area in Building

C-410, these levels are consistent with what one would expect to find from surface deposition in the general area.

- C No radiation levels (greater than 5 mrem/hr) were found on direct readings of the gold slag, molds, crucibles, or other equipment.

A health physics technician at the site alleged that he once surveyed a crucible stored in C-746A that was stamped "C-400." The survey revealed radiation readings of several hundred mrad per hour. The technician believed that this crucible was used for melting gold. The technician toured C-746A with a representative of the team. The crucible could not be found. The team requested information on the whereabouts of this specific crucible in a bulletin that went to all Bechtel Jacobs and USEC employees at the PGDP site. No one came forth with any information about the crucible. Therefore, the allegation of high radiation readings on that particular crucible could not be substantiated.

- C Gold slag believed to be from the gold recovery process contained very low levels of uranium contamination.

A gold slag sample (**Exhibit 6**) believed to be from the C-400 gold recovery operation contained very low levels of radioactivity. Although the origin cannot be proven, this gold slag was given to the investigation team by an engineer involved in the gold recovery program. The slag was analyzed by mass spectrometry and gamma spectroscopy. All isotopes except ^{238}U were less than their respective minimum detectable activities (MDAs). Concentrations of the ^{238}U were low (8.65 pCi/g from gamma spectroscopy on the total sample).

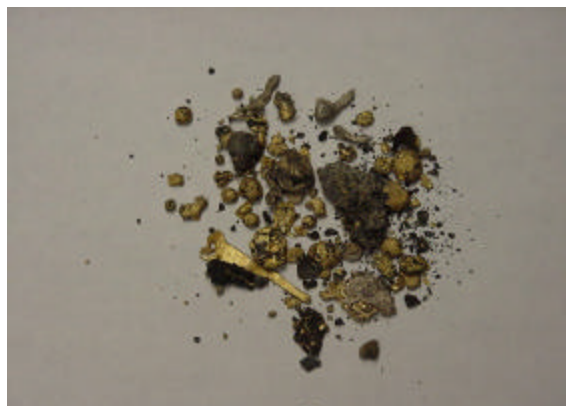


Exhibit 6 - Gold slag.

3.2.5 Potential For Release of Contaminated Material

- There was a potential for cross-contamination of the gold with uranium, technetium-99, and neptunium-237 during recovery operations in Building C-400.

Because the gold was recovered and melted in C-400, one of the more highly contaminated buildings at PGDP, there was potential for cross-contamination of the gold ingots produced there. Drums of gold-plated weapons parts received after disassembly in C-746A were stored in what is now referred to as the Neptunium Recovery Room of C-400 (**Exhibit 7**). Surveys for removable contamination on items stored in the room for many years were recently conducted. Levels were below DOE's unrestricted use limits for beta emitters and only slightly above the unrestricted use limits for transuranics. Since

the gold was in drums, it apparently did not come



Exhibit 7 - Neptunium Recovery Room.

into direct contact with the contamination while in storage.

The major steps in the gold recovery operation, including melting the recovered gold flakes into bars, were performed in the west high bay of C-400. The gold scrap dissolver and furnace doors were closed during operation. The gold hand-table was

open to the C-400 high bay (**Exhibit 8**), which is a contamination area by today's definition. Surveys were conducted on surfaces of the scrap dissolver, furnace, and hand-table that would have the highest potential for contacting or cross-contaminating the gold. While contamination was detectable, levels generally met DOE unrestricted use limits, even after years of being stored in contamination areas.

The potential existed for cross-contamination of the gold. The uranium and ^{99}Tc in the solid residues on the gold crucibles are the strongest indicator that there may have been a cross-contamination problem with the gold itself. In a comparison of contamination levels from the used crucible with those of the unused (control)

crucible, those from the used crucible seem to exceed what would be expected from surface deposition during storage. However, whether or not this contamination was present at the time of the gold melting is uncertain.

3.3 SILVER

3.3.1 Material Processing Description

- Film was incinerated in C-405, the ash was smelted in the C-727 Foundry, and the resulting silver bars were sold.



Exhibit 8 - C-400 high bay area.

According to interviews with former employees who worked in the silver process, the silver-bearing film was stored in the gold storage area of the C-746 Building. The film was delivered to the C-405 incinerator for burning in lots of approximately 5,000 lbs. After burning, the ash was collected in 5-gallon containers and delivered to the C-727 Foundry (**Exhibit 9**). The ash was mixed with 20# mule team borax and smelted, and silver bars were produced. Standard Operating Procedure CH-359, *Silver Recovery in C-400*, was obtained through the Department of Justice. The procedure is dated April 28, 1978, and states that the silver

recovery took place in Building C-400. This is in direct conflict with the information obtained from former employees and the date of the procedure is long after the end of the silver recovery process. No one at the site could explain the discrepancies.

- Paducah ceased production of silver bars in 1974 but continued shredding/burning film and selling the silver-bearing ash.

After 1974, Paducah ceased recovering silver but continued shredding and/or burning the film. This may have coincided with the closure of the C-727 Foundry. The film ash or shredded film was analyzed by the onsite C-710 Laboratory for the silver content and the product was sold to the highest bidder. The buyer was then responsible for recovering the silver. Per the C-746 Building Smelter Production Logbook, Volume 5, beginning in 1979, starting with 13,000 lbs. of film on-hand and continuing through 1985, a total of 173,372 lbs. of film were received at Paducah. The film was received from Rockwell at Rocky Flats, Bendix, General Electric, onsite at Paducah, Pantex, Portsmouth, and miscellaneous reproduction and photo labs. A letter dated December 2, 1985, indicates that an unexpected policy change by DOE restricted the scope of



Exhibit 9 - C-727 Foundry.

silver recovery services that Paducah could perform. Records indicate at that time Paducah returned service orders since they were unable to perform the burning services requested. Records indicate that on April 15, 1986, Paducah sold shredded scrap film into commerce. No other records were found after that date. Procurement records for the sale of the silver-bearing ash could not be located by PGDP or by the team.

3.3.2 Quantities of Material Recovered

- According to site records, beginning April 28, 1966, until May 2, 1974, 102 silver bars were produced on site (more than 7,600 pounds).

Records indicate that the silver was recovered from scrap received (predominantly x-ray film) from Burlington/Pantex, Dow Chemical, and Bendix. However, records for 21 of the poured bars were not found nor could PGDP produce them. It is not known from which facility(ies) the film scrap for these 21 bars were received. The silver logbook records for the remaining 81 bars indicate approximately 7,650 lbs. of silver were recovered with a total revenue of around \$315,000.

3.3.3 Disposition of Material

No procurement records could be produced to indicate the quantity of silver sold nor the selling procedure. Records reviewed from the U. S. Treasury did indicate that some silver from Paducah was processed at the New York Assay Office, but no information was available to indicate what was done with the material.



Exhibit 10 - Nickel ingots.

3.3.4 Analytical Results

- Analysis of C-405 incinerator ash composited from 28 spools of film revealed no radioactive contamination.

A composite ash containing approximately 3% silver was found in the C-710 Laboratory and analyzed at Oak Ridge National Laboratory (ORNL) by ICP mass spectrometry. All isotopes were below the detection limits for the analysis.

- A bucket of silver slag found in the C-400 Neptunium Recovery Room was analyzed by ICP mass spectrometry and found to contain 2,100 pCi/g of technetium-99.

From the silver slag found in the bucket, ^{99}Tc was the only isotope that exceeded levels one would associate with natural background. The slag was first given an acid wash to remove external contamination; the acid wash showed no ^{99}Tc above the MDA. Therefore, the slag is believed to be at least partly internally contaminated. From interviews, a previous production manager indicated that this slag was not from film but from

tests on the gold dissolver sludge. The sludge did contain significant quantities of silver. An economical method for recovery was never found. Efforts to sell the sludge for its silver content were also unsuccessful.

3.3.5 Potential for Release of Contaminated Material

There was a potential for cross-contamination of the silver. There is no evidence to indicate a potential for contamination at a level that would have public health consequences.

3.4 NICKEL

3.4.1 Material Processing Description

- Operation of a nickel barrier manufacturing facility and operation and maintenance of the gaseous diffusion plants resulted in several sources of scrap nickel.

Gaseous diffusion barrier is manufactured from sintered nickel powder. Nickel scrap not suitable for barrier manufacture was recycled. The only method deemed adequate for destruction was to smelt the material into a solid block or ingot (**Exhibit 10**). Assuming no cross-contamination occurred within C-746A, these ingots were free of radioactive contamination since the metal was never exposed to the diffusion process gas. During the manufacturing process for diffusion barrier, incomplete and flawed barrier and barrier scrap resulted. These scrap materials were classified, and so destruction by smelting was required. Both the scrap and resulting ingot were free of radioactive contamination, assuming no cross-contamination occurred.

During process operations, individual barrier tubes failed. These tubes were removed from service. Scrap barrier from the barrier removal activity had been exposed to process gas and was therefore, radiologically contaminated. Similarly, the diffusion barrier was replaced at the Paducah, Portsmouth, and selected process buildings at the K-25 plant during the CIP/CUP activities. The removed barrier was crushed, shipped to Paducah, and smelted. Although classification concerns were removed, the resulting ingots were radiologically contaminated since the material was not completely decontaminated prior to smelting. The principle radionuclides of concern are uranium (^{234}U , ^{235}U , ^{238}U), ^{99}Tc , and trace quantities of ^{237}Np and ^{239}Pu .

3.4.2 Quantities of Material Recovered

- Clean nickel was processed through the smelter before the contaminated nickel. Approximately 17 million lbs. of nickel were smelted, cast into ingots, and sold.

The operating records for the C-746A Smelter indicate that “clean” nickel (not contaminated with radiological materials above the release criteria) was smelted into ingots. A chart detailing amounts of product sold (including aluminum, monel, cobalt, and lead) is included in **Attachment 5**.

- Contaminated nickel (volumetrically contaminated with radiological materials) was processed through the smelter from December 1982 to January 1986. Approximately 19.6 million lbs. of contaminated nickel were cast into ingots during this period.

As each ingot was poured, a sample button was

cast (**Exhibit 11**). These nickel buttons were analyzed for elemental and radionuclide composition. Each ingot was embossed and each button was given a unique identifier that cross-referenced each ingot to the analytical results from each button. None of the known contaminated nickel material was sold to private industry or otherwise released from radiological control. The material is currently maintained in the Paducah contaminated scrap yard.

3.4.3 Disposition of Material

The nickel campaign ran from mid-1976 to late 1982. The “clean” nickel ingots were sold into commerce between 1976 and 1983 (and one sale in 1991 of 1,820 lbs.). The contaminated



Exhibit 11 - Nickel and aluminum sample buttons.

nickel ingots remain on plant site.

3.4.4 Analytical Results

- There was some potential for cross-contamination of clean nickel ingots that were produced in C-746A subsequent to the introduction of contaminated barrier to the induction furnace.

Both clean (unused) and contaminated nickel barrier were melted and staged in C-746A.

Although most of the clean nickel was melted prior to the introduction of contaminated barrier to the induction furnace, some clean nickel was melted after contaminated material had been introduced. The induction furnace liner was changed between processing campaigns of contaminated and clean nickel, so the potential for cross-contamination of clean nickel from the furnace was very remote. However, this would not preclude the possibility of the clean nickel ingots becoming cross-contaminated by their handling and staging in the general area of the furnace. Although isotopic analyses were not conducted on surface contamination smears in the vicinity of the furnace, uranium, ^{99}Tc , and ^{237}Np contamination were potentially present due to the melting of the contaminated nickel barrier in previous processing campaigns. Area and breathing zone air samples taken during the contaminated nickel melts consistently showed uranium, ^{237}Np , and ^{99}Tc . However, based on a January 29, 1999, survey, removable surface contamination levels on and around the induction furnace generally met current DOE free-release limits even when transuranic release limits were applied. Other than floor sweeping, the facility does not appear to have been decontaminated or painted since the induction furnace was shut down.

- C As of 1983 and possibly sooner, procedures required contamination surveys of nickel ingots that were produced in C-746A and subsequently sold. The procedure required documentation of these surveys; however, no documents were found.

3.4.5 Potential for Release of Contaminated Material

- C Analyses of randomly selected samples from

nickel ingots sold into commerce indicate the presence of contamination above normal background levels.

Nickel sample buttons obtained during the ingot pours have been analyzed by mass spectrometry. The buttons were volumetrically contaminated with ^{99}Tc at levels up to 23,500 pCi/g and ^{239}Pu at levels up to 7.5 pCi/g.

3.5 ALUMINUM

3.5.1 Material Processing Description

Aluminum was recovered at the PGDP from 1967 to 1986. Two aluminum melting furnaces located in Building C-746A were used for this process. The furnaces were used to melt classified aluminum scrap (clean and contaminated) received from various locations. Melting was necessary to destroy the classified nature of the material. The Sweat Furnace was used solely to melt clean aluminum scrap and the Reverberatory Furnace was used to melt contaminated aluminum scrap. Standard Operating Procedure U-82, *Operation of C-746A Aluminum Melting Furnaces*, dated July 20, 1978, describes the process of aluminum recovery. As the ingots were produced, "button" samples were poured.

3.5.2 Quantities of Material Recovered

From 1970 until 1986, approximately 4.5 million lbs. of recovered aluminum was processed into ingots and sold into commerce.

3.5.3 Disposition of Material

The aluminum ingots determined to be "clean" (not contaminated with radiological materials above the release criteria) were released into

commerce. Contaminated aluminum ingots currently remain onsite at the Paducah GDP.

3.5.4 Analytical Results

- C Incoming aluminum weapons components were not surveyed at PGDP until 1982.
- C Procedures required contamination surveys of aluminum ingots produced in C-746A and subsequently sold. The procedure required documentation of these surveys, but no documents were found.
- C There is radio chemical data on aluminum that was sold after 1984.

PGDP analytical results were used to determine contamination levels and to segregate aluminum containing enriched uranium from natural and depleted uranium. Aluminum containing enriched uranium or containing greater than 50 ppm total uranium was not sold after 1984. There are records of aluminum analyses and sales only after 1984. The team could not establish if aluminum was analyzed or sold prior to that time.

3.5.5 Potential for Release of Contaminated Material

- C There was potential for cross-contamination of aluminum ingots that were produced in C-746A.

Clean aluminum was melted and staged in C-746A. The potential for cross-contamination of aluminum ingots was low given that the aluminum furnace was isolated from the processing of the contaminated nickel, the major source of contamination in the building. In recent surveys conducted by Bechtel Jacobs at the aluminum furnaces in C-746A, both removable and total alpha contamination levels met DOE unrestricted

use limits. Removable beta contamination levels also met these limits. Total beta contamination exceeded unrestricted use limits at 27% of the sampling locations. It is not possible to determine what fraction of the total beta contamination was potentially transferrable to the aluminum when the ingots were produced.

- C Randomly selected samples from aluminum ingots that were sold into commerce have been analyzed by ORNL. Actinide concentrations in the samples were considerably in excess of background levels for four isotopes.

Thirty-eight aluminum button samples obtained during ingot pours have been analyzed by ICP mass spectrometry. Thorium-230 (^{230}Th), ^{239}Pu and ^{240}Pu were present in most samples. Natural uranium isotopes were also present in low concentration with total uranium concentrations less than 30 pCi/g. Maximum concentrations for ^{230}Th , ^{239}Pu , ^{240}Pu and ^{233}U were 66 pCi/g, 212 pCi/g, 424 pCi/g, and 18 pCi/g, respectively. (ORNL reports that these numbers are only good to +/-50%.) These values represent the highest actinide concentrations found in any metal.

3.6 STEEL

3.6.1 Material Processing Description

As a result of the CIP/CUP, approximately 26.7 millions lbs. of contaminated steel scrap was generated at PGDP. In an effort to initiate a volume reduction and declassification of the scrap steel, a sample steel test melt was conducted on September

8, 1981 (**Exhibit 12**). Approximately 22,000



Exhibit 12 - Contaminated steel ingots.

lbs. of contaminated steel was melted before termination of the test due to excessive depletion of the furnace liner. Samples from 10 ingots were composited and analyzed for their respective contaminants. These data show that the levels of contaminants are reduced during smelting. However, termination of the project was due to the following problems:

- The induction furnace liner (95% aluminum oxide and 5% magnesium oxide) proved to be incompatible with the slag formed during steel smelting. The slag came from the high iron oxide level covering the surface of the scrap steel; this oxide preferentially attaches to the magnesium oxide, thus accelerating liner erosion.
- An excessive amount of slag would collect at the surface of the molten mass and tend to harden and crust over in cooler areas above the induction coils. The crust formation complicated steel recharging and pouring of ingots. The crust had to be manually broken and removed creating potential safety problems.
- The collection of slag at the surface of the molten mass increased the possibility of
- contaminant carryover into the ingots.

The ingots produced during the test melt and the

remainder of the contaminated steel scrap remain at Paducah in the contaminated scrap yards.

3.6.2 Quantities of Material Recovered

Approximately 26.7 million lbs. of contaminated steel scrap was generated at PGDP. The total quantity of clean steel sold since the Plant's construction is unknown.

3.6.3 Disposition of Material

Excess clean steel has been sold into commerce from Paducah since the Plant's construction. The steel scrap was placed in the clean scrap yard until a successful bidder purchased a supply.

The Operating Contractor at Paducah has routinely accumulated and sold uncontaminated scrap metal. The scrap metal resulted from maintenance and renovation activities. Two approaches have been used at ORO facilities to determine that the scrap metal was uncontaminated and therefore available for public sale and recycle: (1) For scrap metal arising from facilities without any presence or history of housing or processing radioactive materials, process knowledge was the basis for the uncontaminated declaration. A sample of the accumulated scrap metal determined to be uncontaminated was reported to have been verified as uncontaminated by health physics measurement prior to sales. (2) For scrap metal arising from facilities with a known presence or history of housing or processing

radioactive materials, discrete health physics measurements were used for the basis of the uncontaminated declaration.

The ingots produced during the test melt and the remainder of the contaminated steel scrap remain

at Paducah in the contaminated scrap yards.

- In July 1991, DOE imposed a temporary moratorium on the sale and disposal of materials. Following the moratorium, the process of accumulating scrap metal for public sale was changed. Presently, scrap metal that has been determined to be suitable for public recycle is accumulated in 1000 ft³ storage containers in access-controlled areas.

3.6.4 Analytical Results

Since the recovered steel was deemed “clean,” no formal surveys were performed on the material. However, interviews with health physics staff indicate that routine “spot checks” were performed for radioactivity and occasionally radioactive contaminated material was found. When contamination was identified, the material was moved to the contaminated scrap yard. No documentation was available to substantiate these checks.

3.6.5 Potential for Release of Contaminated Material

The potential for release into commerce appears to have been small. However, there is no documentation to substantiate that only clean material was released throughout the history of the PGDP.

3.7 COPPER

3.7.1 Material Processing Description

As a result of the CIP/CUP project at the PGDP, a considerable amount of copper and copper-containing parts were accumulated. The copper

that was salvaged was shipped to NLO, Inc. (Fernald Plant) for storage, smelting, and/or other volume reduction in 1984 at DOE’s direction. NLO was instructed to transfer the processing costs and copper shredder improvements to Paducah (the costs were transferred on January 11, 1985). There is no evidence to indicate that any copper was sold into commerce from Paducah prior to the transfer to the Fernald Plant.

3.7.2 Quantities of Material Recovered

Advances in automation and handling techniques have resulted in a reevaluation of the economics of processing, monitoring and releasing this copper. As a result, all of the Fernald Plant copper inventory (from all three GDP’s) has been shipped to the ETTP in Oak Ridge, Tennessee, for recovery under an Oak Ridge Operations contract with Decon and Recovery Services, Inc. (DRS). The process is near completion with all of the recovered copper being sold to date (using surface release criteria specified by DOE Order 5400.5).

As of June 27, 2000, approximately 2,007,790 lbs. of copper has been recovered by DRS and sold.

3.7.3 Disposition of Material

The recovered copper was being released into commerce before the Secretary of Energy suspended all metals recycling activities.

3.7.4 Analytical Results

As part of the Work Package, DRS performs rad surveys of a percentage of each shipment, and then an independent verification is performed by Oak Ridge Institute for Science and Education.

3.7.5 Potential for Release of Contaminated Material

There is no evidence that copper was released into commerce until recently. Releases had been made in compliance with DOE Order 5400.5 until the DOE suspension of metals recycling activities.

3.8 COBALT/MONEL

3.8.1 Material Processing Description

In addition to the relatively large quantities of steel, nickel, and aluminum, the ORO scrap metal facilities have recycled small quantities of other metals; e.g., monel and cobalt (**Exhibit 13**). The major process source of the monel (a copper-nickel alloy) was the manufacture of new barrier. Monel was used to fabricate the ferrules of the barrier tube. Off-specification monel feedstock and monel recovered from off-specification barrier tubes were accumulated, smelted to destroy any classification concerns, and sold for recycle. Cobalt was used as a consumable component of processes at the K-25 Plant (now ETTP).

Although these metal streams were associated with the manufacture of new (uncontaminated) barrier and other processes, during review of “Operating Instructions (to be used for temporary Standard Operating Procedures)” that pertained to C-746A operations, records showed that on April 29, 1986, a sintering program for a monel-nickel run was started. It indicated that eight 55-gallon drums of monel were mixed with one drum of contaminated nickel pellets. This information corresponds to the Production Logbook which indicated that on April 29, 1986, there was a production run of monel, which was subsequently sold. There were no indications in the logbook



Exhibit 13 - Cobalt ingot.

that this monel was mixed with contaminated nickel.

3.8.2 Quantities of Material Recovered

Review of the PGDP Production Logbook indicates that 882,440 lbs. of monel were released into commerce as well as 20,800 lbs. of cobalt.

3.8.3 Disposition of Material

Monel and cobalt were sold into commerce in the amounts listed above from 1981 through 1985. Also, in the Production Logbook under cobalt was the notation that 50,700 lbs. of cobalt were “shipped to Oak Ridge.”

3.8.4 Analytical Results

None available.

3.8.5 Potential for Release of Contaminated Material

There is evidence that contaminated monel-nickel was potentially released into commerce. There is no evidence that the contamination was at a level that would have public health consequences.

potential for contamination levels that would jeopardize the public.

3.9 LEAD

3.9.1 Material Processing Description

Lead was received and recovered from several sources, including weapons parts. The team found only information on the sale of shredded lead. The use of lead in “outside work” is covered in another report.

3.9.2 Quantities of Material Recovered

The C-746A Smelter Production Logbooks indicate approximately 258,990 lbs. of shredded lead were produced and sold into commerce.

3.9.3 Disposition of Material

The records indicate that the shredded lead was sold into commerce. No information was available on where in commerce this material went.

3.9.4 Analytical Results

None available.

3.9.5 Potential for Release of Contaminated Material

The shredded lead was listed in the C-746A Smelter Production Logbooks; therefore, there was a small potential for cross-contamination. However, the team found no evidence to indicate

4.0 RISK ASSESSMENT

4.1 Gold

Auxier and Associates was contracted to provide a worst-case risk assessment for recovered gold. This assessment was based upon the use of gold as an injected anti-arthritis agent. This worst-case analysis of potential radiation doses associated with recycled PGDP gold indicates that any doses from the injections would have been small. It is quite unlikely that doses in excess of about a tenth of the annual natural background would have occurred if the recycled gold had been used directly without admixture to refining. These conclusions are largely based on the analysis of process residues and are subject to revision if some of the actual product can be located and analyzed. The full report is included as **Attachment 4**.

4.2 Silver

Public health risks from the release of silver are an order-of-magnitude less than those for gold. This is based on the analyses of the composite film ash and silver slag. The analyses indicated no actinide isotopes. Only ^{99}Tc was present. The highest level was 2100 pCi/g, an innocuous level. A person would have to ingest 80 million pCi of ^{99}Tc to receive even 100 mrem committed effective dose equivalent (CEDE). Based on the very limited samples available, human health risks from the release of the silver would be at least an order-of-magnitude less than those for gold.

4.3 Other Metals

To date, the worst-case analysis performed for gold by Auxier and Associates (direct injection of gold containing Np-237 into arthritic patients) results in the highest exposures among all metals

and all exposure pathways. Therefore, because this worst-case scenario would have resulted in very small dose exposures, the resultant committed dose equivalents to members of the public from the other PGDP recovered metals would have been inconsequential, based upon the data collected for this review.

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5.0 CONCLUSIONS

The potential for cross-contamination of gold transferred to the U. S. Treasury or sold into commerce existed. The uranium and ^{99}Tc in the solid residues on the gold crucibles are the strongest indicator that there may have been a cross-contamination problem with the gold itself. Comparing contamination levels from the used crucible with those of the unused (control) crucible indicate that those from the used crucible seem to exceed what would be expected from surface deposition during storage. However, whether or not this contamination was present at the time of the gold melting is uncertain.

While the team did find indications of cross-contamination of the silver, aluminum and nickel, there was no evidence to indicate a potential for cross-contamination at a level that would have had public health consequences. Even aluminum, which had the highest actinide concentrations of the metals, would not have presented a significant health hazard for the general public. The team however, cannot conclude at this time whether remelting of the aluminum might have posed some exposure risk to foundry workers processing the aluminum.

Copper was released into commerce using surface release criteria specified by DOE 5400.5 prior to July 13, 2000, when the Secretary of Energy suspended DOE metals recycling activities.

Records indicate that volume-contaminated steels were not sold. Surface-contaminated steels were sold but were subject to segregation and occasional verification surface scans by PGDP Health Physics. However, no documentation exists to substantiate these scans.

There is evidence that monel was mixed with contaminated nickel on one occasion and potentially released into commerce. No record exists on the contaminant level of the nickel, thus it is impossible to calculate the level in the monel.

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6.0 TEAM MEMBERS AND STAFF

David Allen, Technical Support Division, Team Lead

Antonio Acuna, Procurement and Contracts Division, Member (retired 12/99)

Andy Bassett, Technical Support Division, Member

Richard Meehan, Facilities and Materials Reuse Division, Member

Jenny Mullins, Technical Support Division, Member

Advisors to the Team:

Richard Martin, Technical Support Division

Timothy Joseph, Technical Support Division

Larry Sparks, Derivative Classifier

Administrative Support:

Melisa Hart, Critique, Inc.

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ATTACHMENT 1

DOE METALS RECYCLING PROGRAM INVESTIGATION TEAM

A Team from DOE/Oak Ridge has been onsite since the end of August working to formulate a historical picture of Paducah's Metals Recycling Program (includes gold, silver, aluminum, nickel, cobalt, monel, tantalum, steel, copper and lead). While a lot of documentation has been gathered and a number of employees (past and present) interviewed, several gaps still exist. The Team is requesting anyone with knowledge of the following to please make contact.

- Documentation or specific knowledge of the silver recovery process involving the pouring of silver bars which occurred from 1966 to 1974
- Standard Operating Procedure CH-359, Silver Recovery Process
- Radiochemical analysis of any metals that were sold
- Gold wash table (used in C-400) and the gold crucible that used to be stored in C-746A
- Gold or silver samples that may still be available
- Perpetual inventories and accountability records of precious metals
- Records that indicate customers (incoming and outgoing) for precious metals

If you or anyone you know has knowledge that may be helpful to the Investigation Team, please call:

David Allen, 423-576-0411 (cell phone: 423-599-8514)

Jenny Mullins, 423-576-0836 (Oak Ridge) or 441-5381 (Kevil)

ATTACHMENT 2

Attachment 2

Gamma Spectroscopy and Mass Spectrometry Results for PGDP Metals Samples and Residues

Attachment 2.1, Gamma Spectroscopy Results from Gold Slag Analysis.

Attachment 2.2, Mass Spectrometry Results for Gold Slag, Gold Process Residues, Silver Slag, and Photographic Film Ash.

Attachment 2.3, Photographs of Gold Slag, Gold Process Residues, Silver Slag, and Photographic Film Ash.

Attachment 2.4, *Preliminary Analytical Results for Nickel Button Samples. The buttons with the positive technetium-99 results are currently being decontaminated and resampled in order to distinguish surface from volume contamination.*

Attachment 2.5, Analytical Results for Aluminum Button Samples (TBD). *Due to classification issues, analysis of the aluminum buttons has been delayed. The buttons are scheduled for delivery to Oak Ridge National Laboratory by mid-July, and the analyses should be completed by mid-August 2000.*

Attachment 2.1
Gamma Spectroscopy Results from Gold Slag Analysis

Nuclide	Concentration (ng/g)	Activity Concentration (pCi/g)	Relative Error
²³² Th	1.6E3	1.8E-1	29 %
²³⁵ U	7.6E1	1.6E-1	51 %
²³⁸ U	2.6E4	8.7E0	42%
²³⁷ Np	1.6E-1	1.1E-1	37%

Attachment 2.2

Mass Spectrometry Results for Gold Slag, Gold Process Residues, Silver Slag, and Film Ash

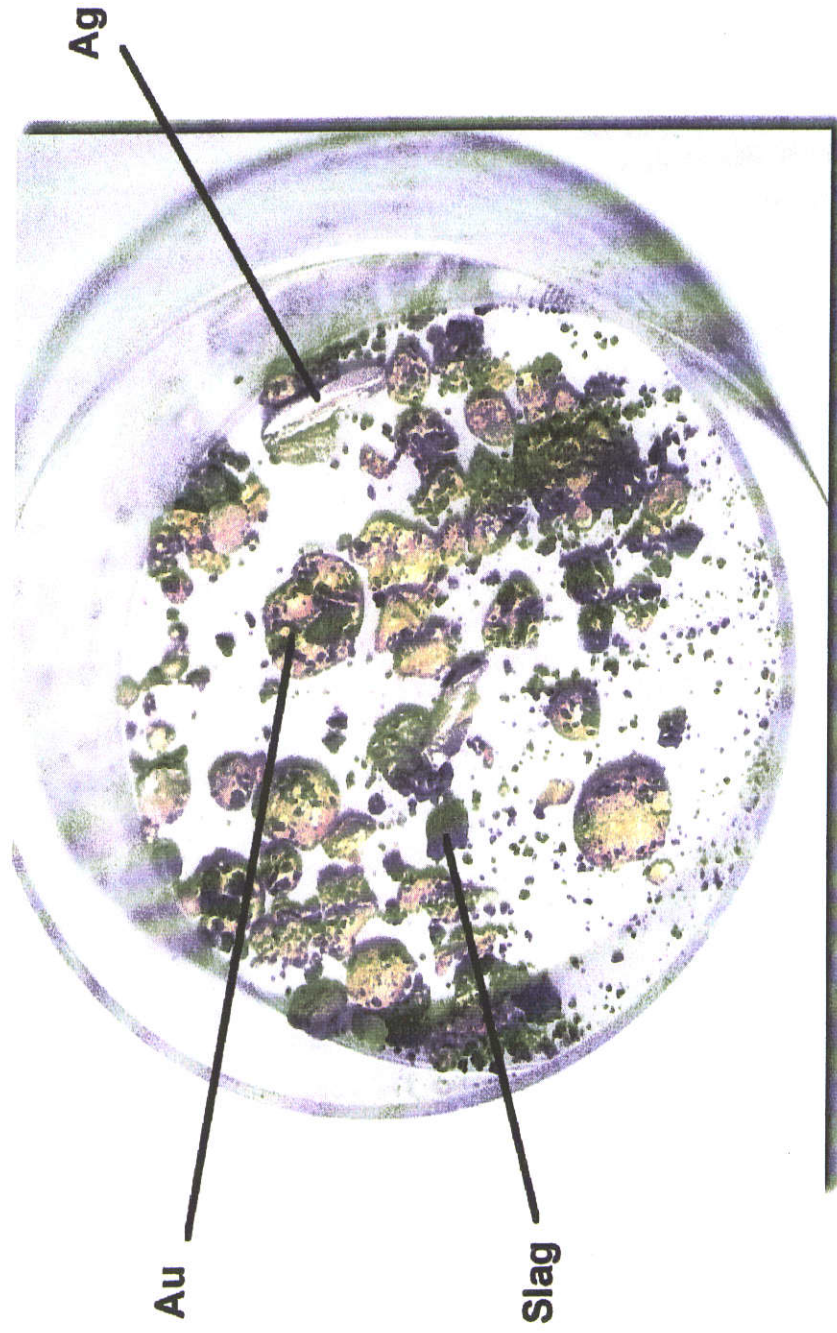
Sample ID	Sample Preparation	Customer Number	RMAL Number	Units	⁹⁹ Tc	²³² Th	²³⁵ U	²³⁸ U	²³⁷ Np
Gold Slag	4M HNO ₃ Leach	990929-1	990929-084	(pCi/g)	< 7.6E+01	< 5.0E-04	< 9.7E-03	4.8E-02	< 3.2E+00
	HNO ₃ -HCL Digest	990929-2	990929-085	(pCi/g)	< 7.6E+02	< 5.0E-03	< 9.7E-02	2.7E-01	< 1.6E+03
Refined Gold	4M HNO ₃ Leach	990929-3	990929-086	(pCi/g)	< 7.0E+00	1.0E-04	2.4E-03	1.4E-01	< 2.9E-01
	HNO ₃ -HCL Digest	990929-4	990929-087	(pCi/g)	< 6.9E+03	< 4.5E-02	< 8.8E-01	1.4E-01	< 1.2E+03
69968 Slag	4M HNO ₃ Leach	990929-5	990929-088	(pCi/g)	< 5.9E+02	2.7E-02	8.2E-02	2.4E+00	< 2.5E+01
	HNO ₃ -HCL Digest	990929-6	990929-089	(pCi/g)	2.1E+03	< 3.9E-03	< 7.6E-02	8.2E-02	< 2.5E+01
Black Crucible Solids 69968	4M HNO ₃ Leach	990929-7	990929-090	(pCi/g)	1.5E+04	1.3E-01	1.0E+01	4.7E+02	< 5.4E+02
	HNO ₃ -HCL Digest	990929-8	990929-091	(pCi/g)	1.7E+04	< 8.4E-02	< 1.6E+00	1.0E+01	< 5.4E+02
Black Crucible Solids 69977	4M HNO ₃ Leach	990929-10	990929-093	(pCi/g)	< 6.6E+03	2.0E-01	1.4E+01	6.4E+02	< 2.8E+02
	HNO ₃ -HCL Digest	990929-11	990929-094	(pCi/g)	1.5E+04	< 4.3E-02	1.9E+00	7.8E+01	< 2.8E+02
Black Crucible 69968	4M HNO ₃ Leach	990929-9	990929-092	(pCi Total)	< 1.7E+03	2.9E-02	2.0E+00	8.4E+01	< 7.0E+01
	4M HNO ₃ Leach	990929-12	990929-095	(pCi Total)	< 1.7E+03	2.9E-02	2.4E+00	1.1E+02	< 7.0E+01
White Crucible Solids 69954	4M HNO ₃ Leach	990929-13	990929-096	(pCi/g)	< 4.5E+03	9.1E-02	1.1E+00	6.6E+01	< 1.9E+02
	HNO ₃ -HCL Digest	990929-14	990929-097	(pCi/g)	< 4.5E+03	3.2E-02	< 5.7E-01	2.4E+00	
White Crucible 69954	4M HNO ₃ Leach	990929-15	990929-098	(pCi Total)	< 1.7E+03	< 1.1E-02	< 2.2E-01	7.7E+00	< 7.0E+01
White Crucible 69982	4M HNO ₃ Leach	990929-16	990929-099	(pCi Total)	< 1.7E+03	1.2E-02	3.6E-01	1.5E+01	< 7.0E+01
Settling Tank	4M HNO ₃ Leach	990929-17	990929-100	(pCi Total)	< 3.4E+04	< 2.2E-01	5.2E+00	2.2E+02	< 1.4E+03
Dissolver Tank	4M HNO ₃ Leach	990929-18	990929-101	(pCi Total)	< 3.4E+04	< 2.2E-01	2.7E+01	8.7E+02	< 1.4E+03
Ceramic Funnel Solids 69969	4M HNO ₃ Leach	990929-19	990929-102	(pCi/g)	< 1.9E+05	< 1.3E+00	< 2.5E+01	1.1E+02	< 8.0E+03
	HNO ₃ -HCL Digest	990929-20	990929-103	(pCi/g)	< 1.9E+05	< 1.3E+00	< 2.5E+01	< 3.8E+00	< 8.0E+03
Acid Evaporator Solids 69975	4M HNO ₃ Leach	990929-21	990929-104	(pCi/g)	< 4.1E+04	< 2.7E-01	< 5.2E+00	1.6E+00	< 1.7E+03
	HNO ₃ -HCL Digest	990929-22	990929-105	(pCi/g)	< 4.5E+04	< 2.7E-01	< 5.2E+00	< 8.1E-01	< 1.7E+03
Acid Evaporator	4M HNO ₃ Leach	990929-23	990929-106	(pCi Total)	< 1.7E+03	< 1.1E-02	< 2.2E-01	5.9E-01	< 7.0E+01
Photo Film Ash	HNO ₃ -HCL Digest	991019-1	991020-124	(pCi/g)	< 6.4E+03	< 4.2E-02	< 8.2E-01	< 1.3E-01	< 2.7E+02
Gold Mold #1	4M HNO ₃ Leach	991026-1	991026-009	(pCi Total)	< 3.4E+03	7.8E-02	2.8E+01	5.3E+02	< 1.4E+02
Gold Mold #2	4M HNO ₃ Leach	991026-2	991026-010	(pCi Total)	< 3.4E+03	6.8E-02	1.4E+01	3.8E+02	< 1.4E+02

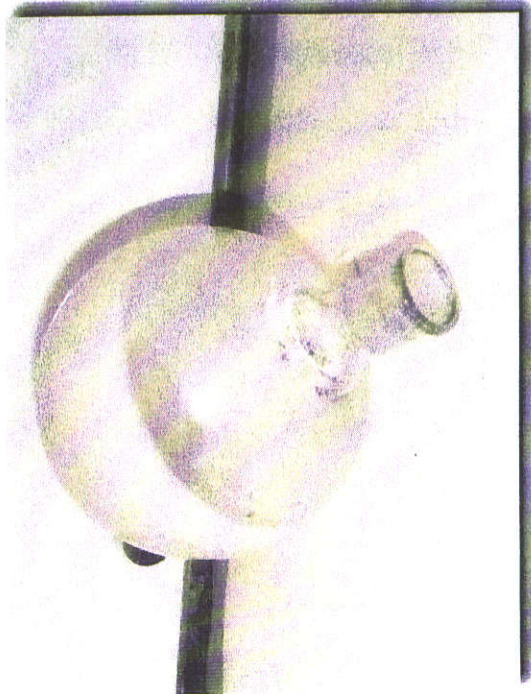
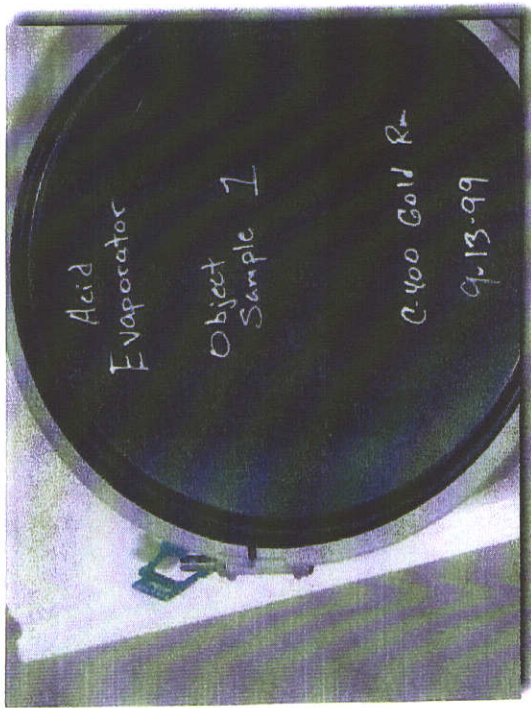
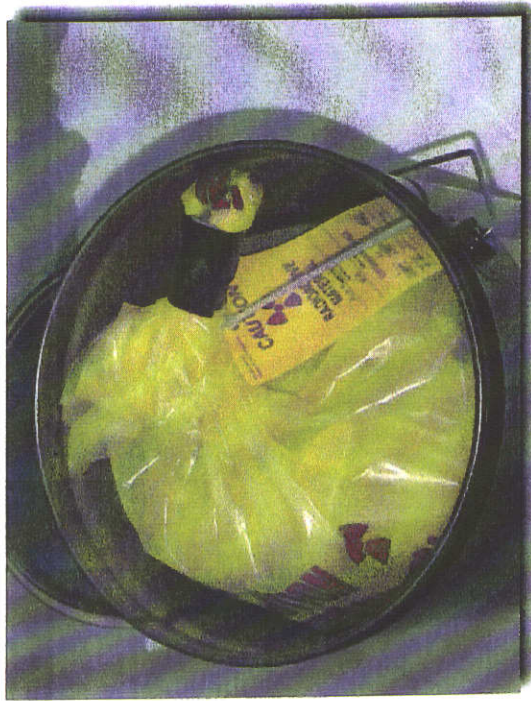
Attachment 2.3

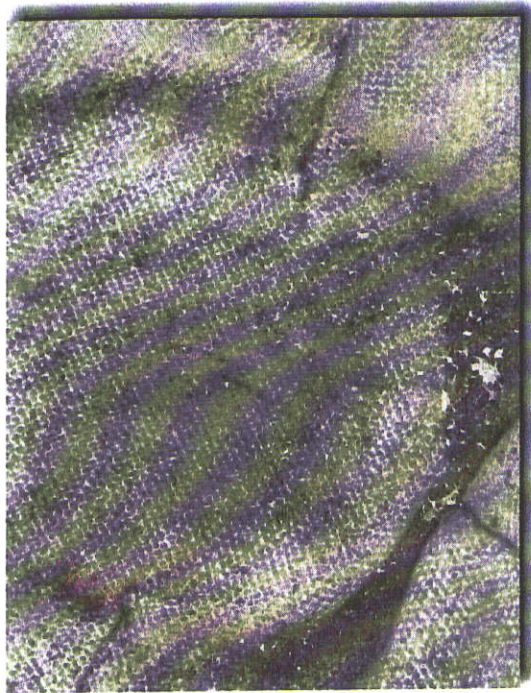
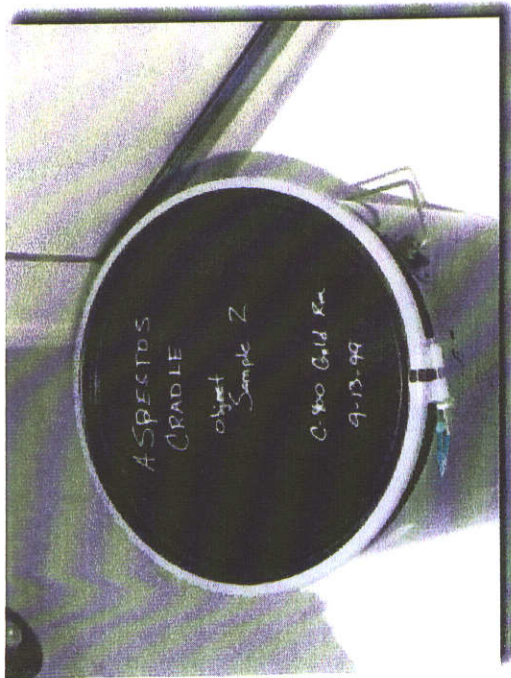
Photographs of Gold Slag, Gold Process Residues, Silver Slag, and Photographic Film Ash

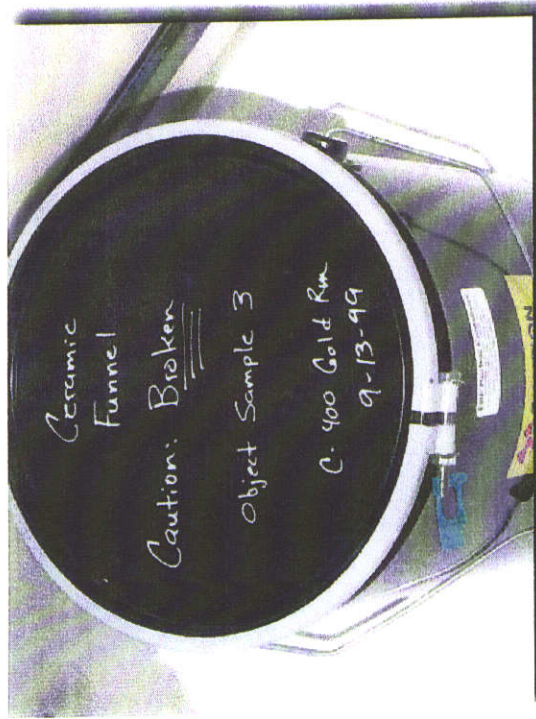
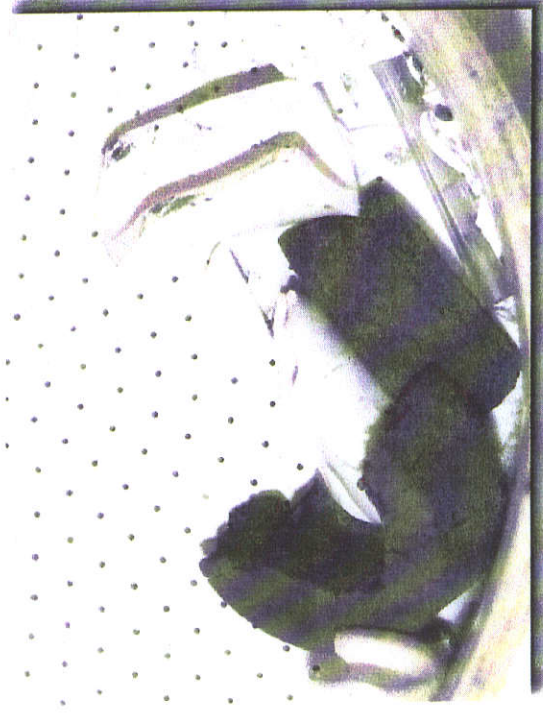
<u>Photographs</u>	<u>Page</u>
Initial Gold Slag sample	1
Sample 1 – Acid Evaporator Round-Bottom Flask	2
Sample 2 – Asbestos Cradle for Flask	3
Sample 3 – Broken Ceramic Funnel	4
Sample 4 – White Ceramic Crucible (Used)	5
Sample 5 – White Ceramic Crucible (Unused)	6
Sample 6 – Black Crucible (#1)	7
Sample 7 – Black Crucible (#2)	8
Sample 8 – Container of Slag	8
Sample 9 – Laboratory Notebook	7
Rad-Swipes from C-400 Gold Room	9
Smear & Wipe Samples from C-400 Gold Dissolver and Settling Tank	9
Gold Mold Parts	10
Gold Furnace and Mold Smears	10
Gold Molds	11
Photographic Film Ash	12

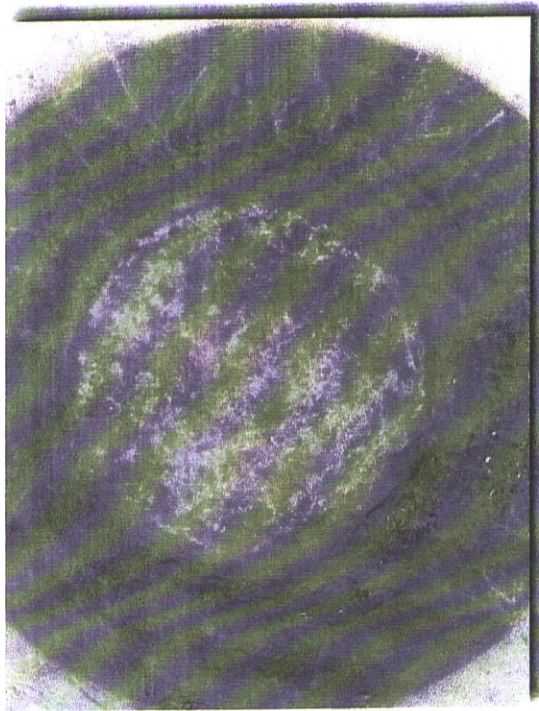
Initial Gold Slag Sample

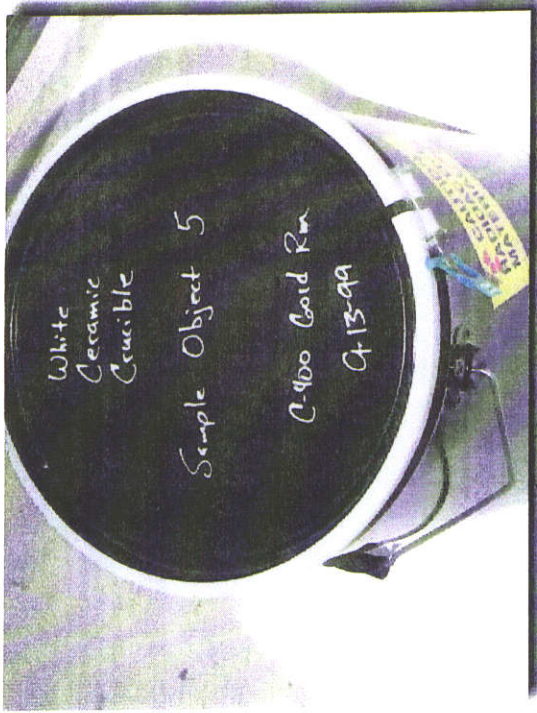




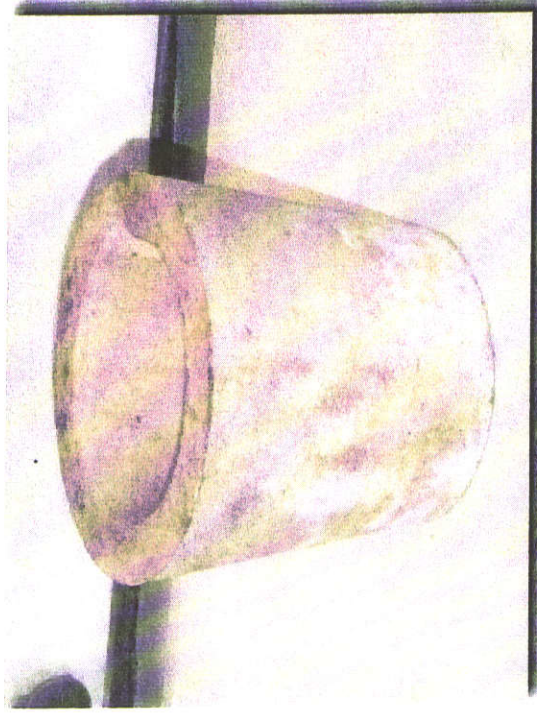
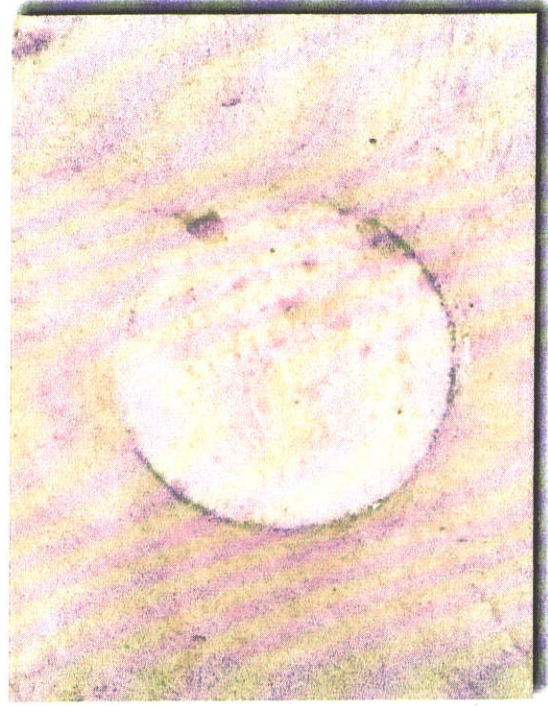


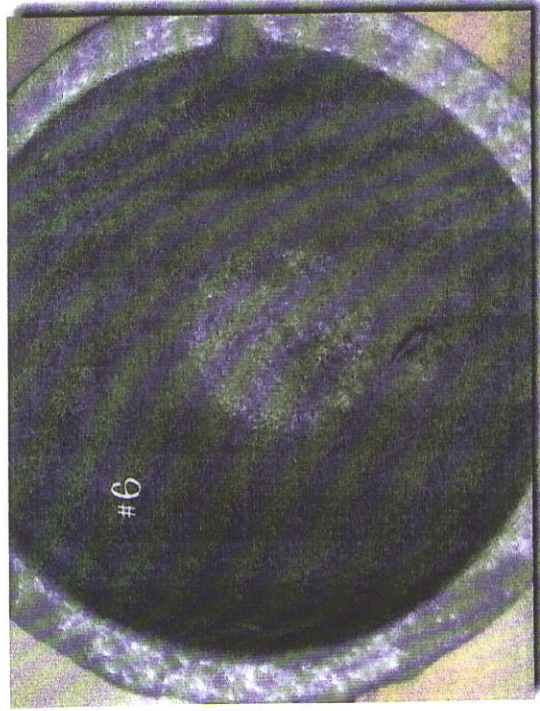
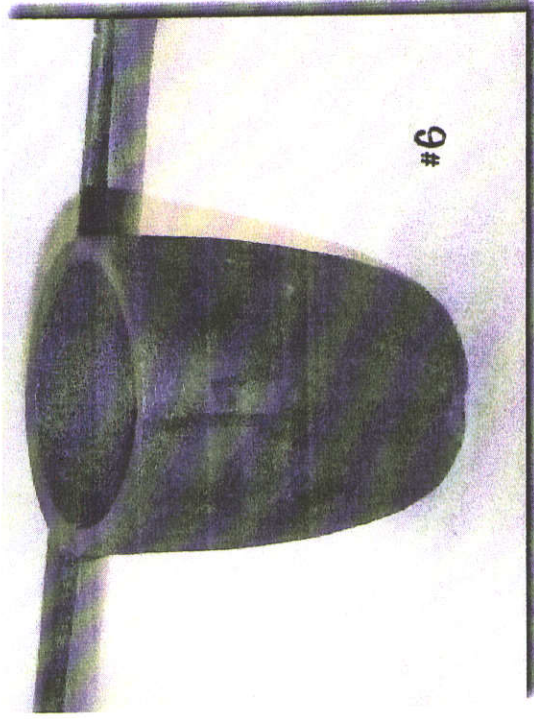
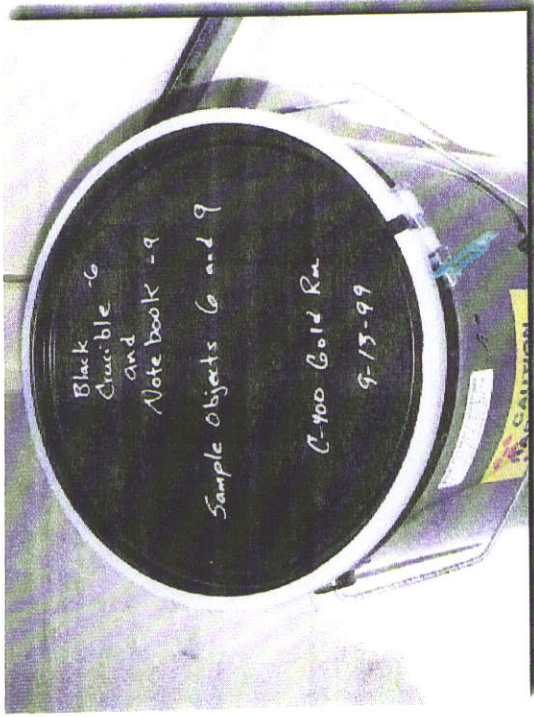


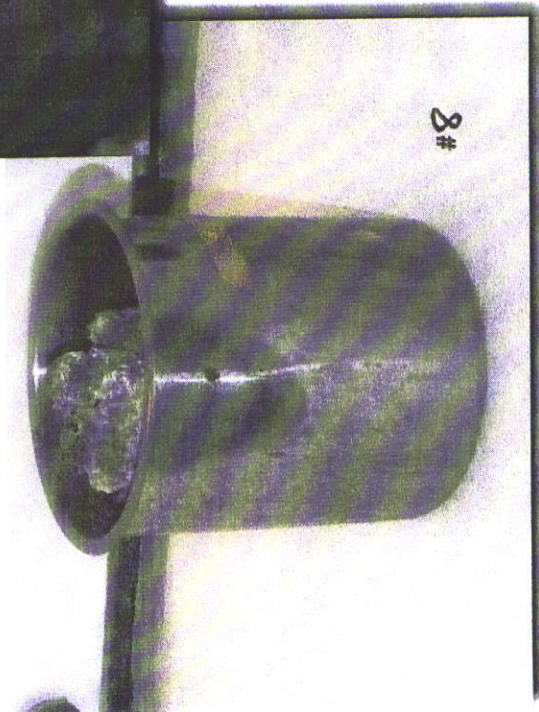
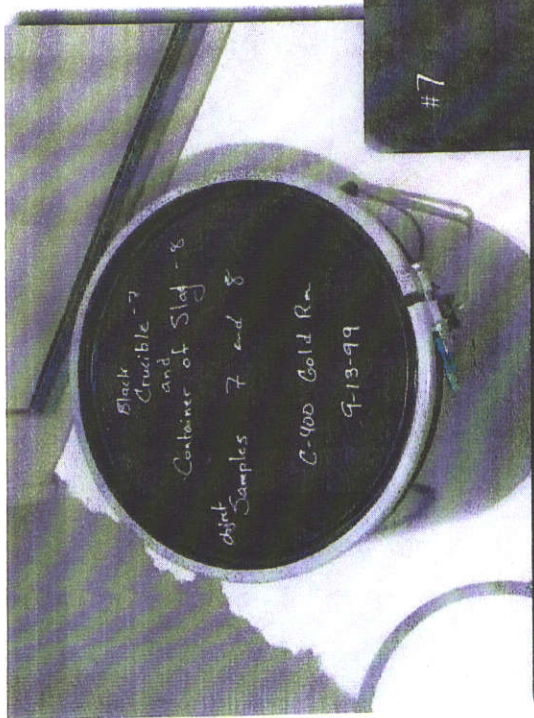
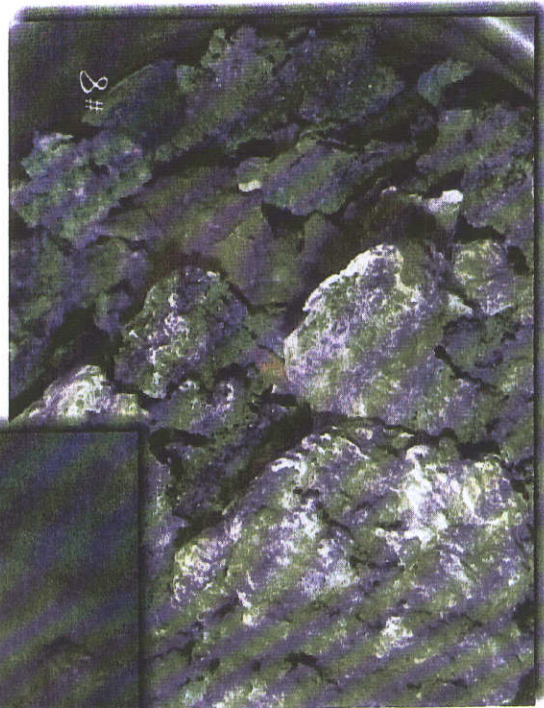
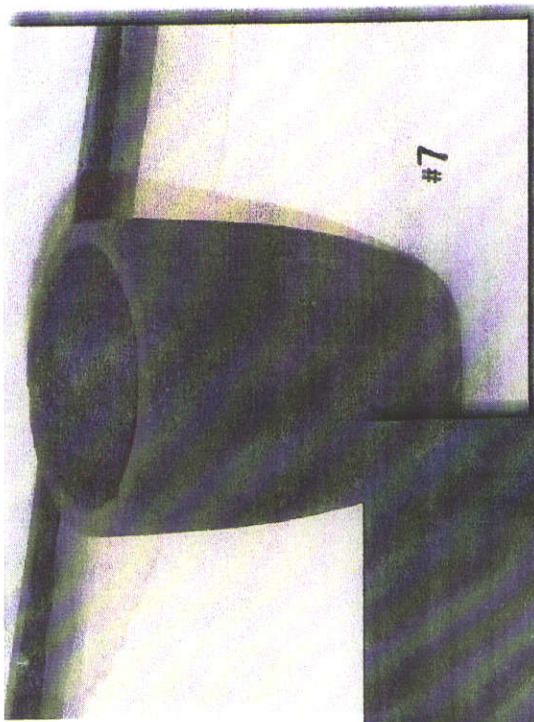


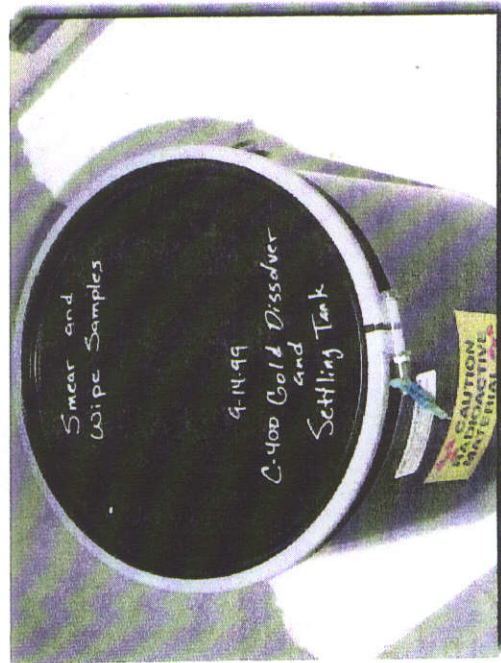
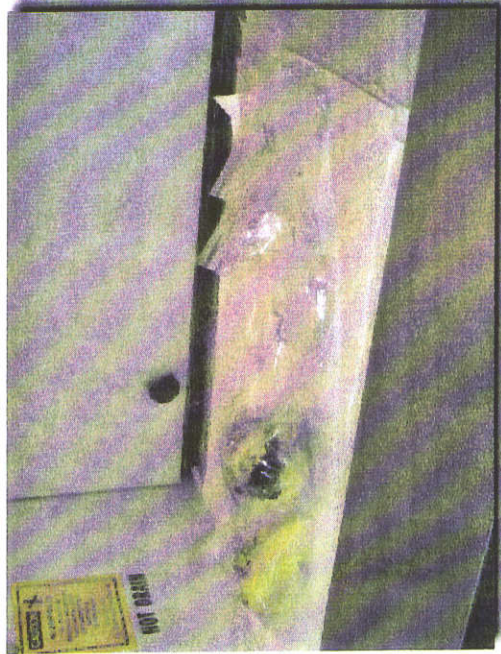


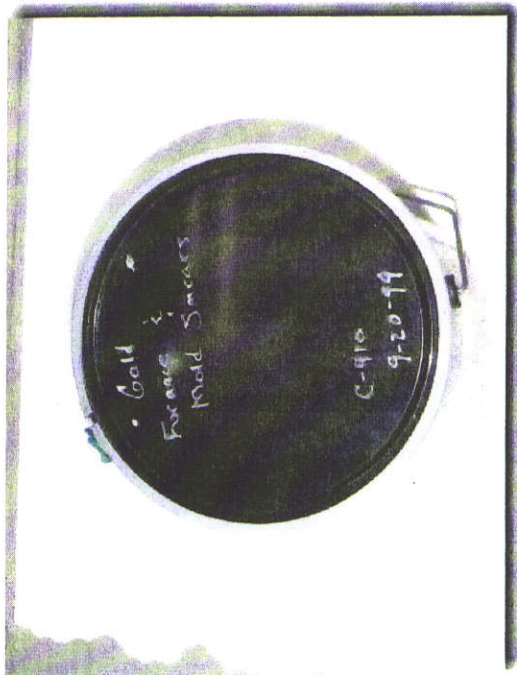
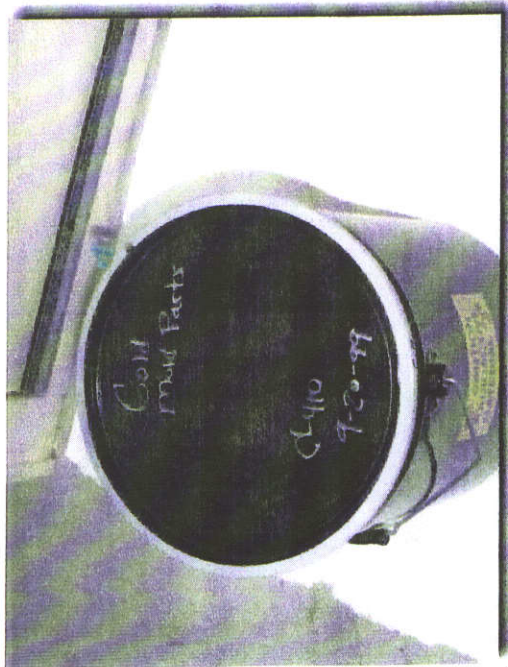
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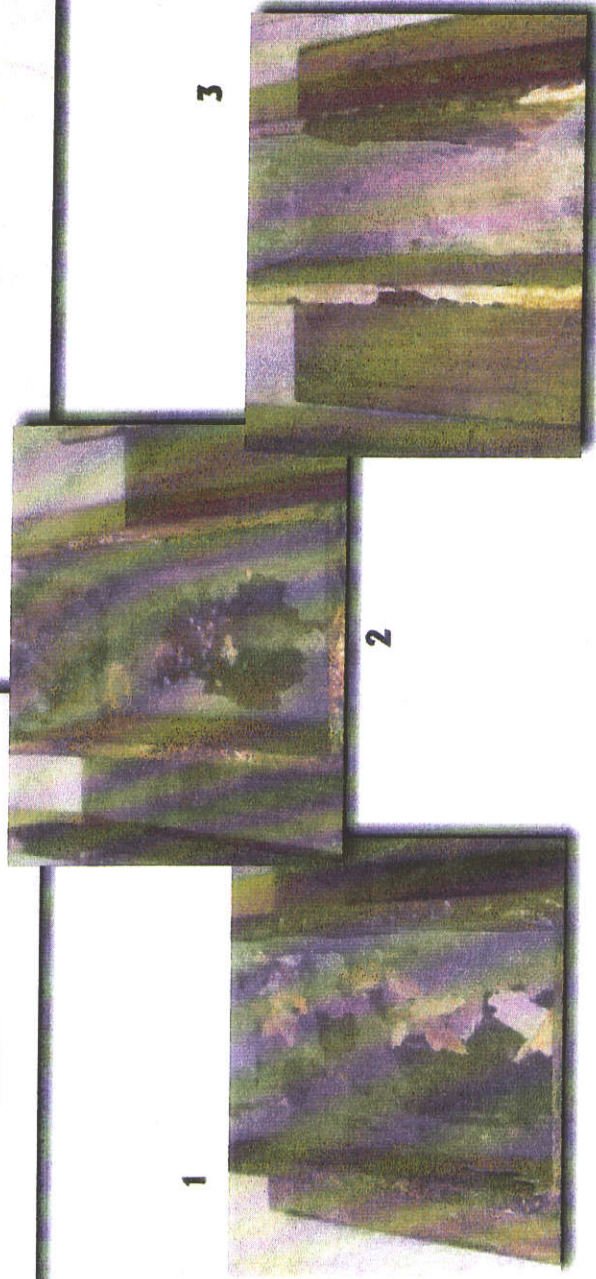


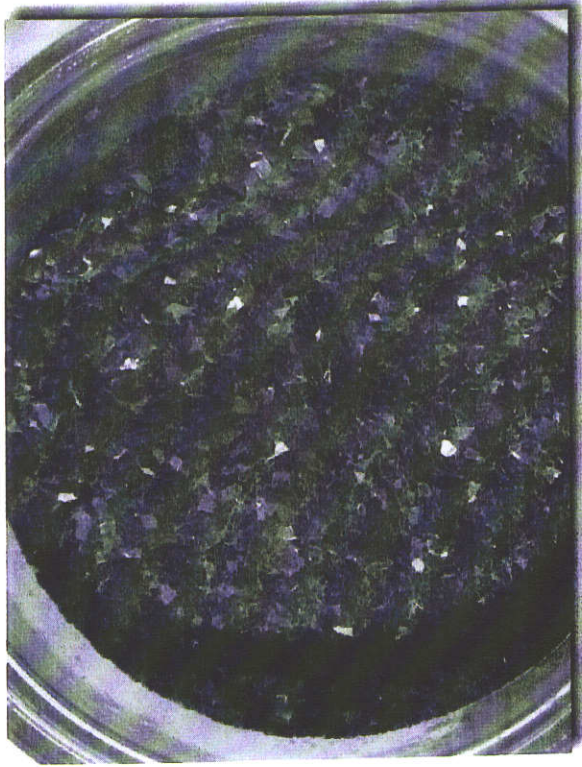












Photographic Film Ash

Attachment 2.4 Preliminary Analytical Results for Nickel Button Samples

RMAL Sample ID	DOE Sample ID	Sample Weight (g)	Th-230 (pCi/g)	Th-232 (pCi/g)	U-233 (pCi/g)	U-234 (pCi/g)	U-235 (pCi/g)	U-238 (pCi/g)	Np-237 (pCi/g)	Pu-238 (pCi/g)	Pu-239 (pCi/g)	Pu-240 (pCi/g)	Pu-242 (pCi/g)	Tc-99 (pCi/g)
000214-012	M127	5.0147	< 1.93E+00	5.84E-05	< 9.00E-01	< 5.82E-01	5.93E-04	3.22E-03	< 6.58E-02	< 1.0E+00	< 5.79E+00	< 2.12E+01	< 3.37E-01	
000214-013	M15	5.4969	< 1.76E+00	3.40E-04	< 8.21E-01	< 5.30E-01	7.10E-04	5.48E-03	< 6.00E-02	< 1.0E+00	< 5.28E+00	< 1.93E+01	< 3.64E-01	
000214-014	M210A	4.8692	< 1.98E+00	1.35E-04	< 9.27E-01	< 5.99E-01	3.58E-04	3.52E-03	< 6.78E-02	< 1.0E+00	< 5.97E+00	< 2.18E+01	< 3.77E-01	
000214-015	M213B	5.0448	< 1.91E+00	1.59E-04	< 8.95E-01	< 5.78E-01	3.41E-04	2.13E-03	< 6.55E-02	< 1.0E+00	< 5.76E+00	< 2.11E+01	< 3.65E-01	
000214-016	M214B	5.1337	< 1.88E+00	9.00E-05	< 8.79E-01	< 5.68E-01	5.18E-04	3.69E-03	< 6.43E-02	< 1.0E+00	< 5.66E+00	< 2.07E+01	< 3.58E-01	
000214-017	M218B	6.5250	< 1.48E+00	1.02E-04	< 6.91E-01	< 4.47E-01	8.97E-04	7.94E-03	< 5.06E-02	< 1.0E+00	< 4.45E+00	< 1.63E+01	< 2.82E-01	
000214-018	M22	6.2923	< 1.53E+00	6.27E-05	< 7.18E-01	< 4.63E-01	6.35E-04	8.75E-03	< 5.25E-02	< 1.0E+00	< 4.62E+00	< 1.69E+01	< 2.92E-01	
000214-019	M232B	5.4361	< 1.93E+00	9.27E-05	< 8.31E-01	< 5.36E-01	1.06E-03	7.18E-03	< 6.07E-02	< 1.0E+00	< 6.06E+00	< 1.96E+01	< 3.38E-01	
000214-020	M272B	5.1742	< 1.87E+00	2.64E-05	< 8.72E-01	< 5.63E-01	8.58E-04	4.39E-03	< 6.38E-02	< 1.0E+00	< 5.61E+00	< 2.05E+01	< 3.55E-01	
000214-021	M28	5.6450	< 1.71E+00	2.05E-04	< 8.00E-01	< 5.17E-01	9.78E-04	9.39E-03	< 5.85E-02	< 1.0E+00	< 5.15E+00	< 1.88E+01	< 3.26E-01	
000214-022	M281B	5.1938	< 1.86E+00	8.92E-04	< 8.69E-01	< 5.61E-01	9.28E-03	2.20E-01	< 6.36E-02	< 1.0E+00	< 5.59E+00	< 2.04E+01	< 3.53E-01	
000214-023	M286A	5.2062	< 1.85E+00	4.23E-05	< 8.67E-01	< 5.60E-01	1.24E-03	1.38E-02	< 6.34E-02	< 1.0E+00	< 6.05E+00	< 2.21E+01	< 3.83E-01	
000214-024	M290A	4.8029	< 2.01E+00	1.56E-04	< 9.40E-01	< 6.07E-01	1.70E-03	3.10E-02	< 6.87E-02	< 1.0E+00	< 5.00E+00	< 1.83E+01	< 3.17E-01	
000214-025	M295B	5.8036	< 1.68E+00	8.00E-05	< 7.78E-01	< 5.02E-01	6.25E-04	1.01E-02	< 5.69E-02	< 1.0E+00	< 5.73E+00	< 2.10E+01	< 3.63E-01	
000214-026	M297B	5.0675	< 1.91E+00	7.35E-05	< 8.91E-01	< 5.75E-01	6.13E-04	8.85E-03	< 5.51E-02	< 1.0E+00	< 5.75E+00	< 2.10E+01	< 3.64E-01	
000214-027	M299B	5.0542	< 1.91E+00	1.15E-04	< 8.93E-01	< 5.77E-01	1.59E-03	3.02E-02	< 6.53E-02	< 1.0E+00	< 5.75E+00	< 2.10E+01	< 3.64E-01	
000214-028	M304B	2.6024	< 2.47E+00	7.05E-05	< 1.18E+00	< 7.47E-01	1.11E-03	1.83E-02	< 8.45E-02	< 1.0E+00	< 7.44E+00	< 2.72E+01	< 4.71E-01	
000214-029	M307A	3.7395	< 2.58E+00	7.17E-05	< 1.21E+00	< 7.80E-01	6.70E-04	1.12E-02	< 8.83E-02	< 1.0E+00	< 7.77E+00	< 2.84E+01	< 4.92E-01	
000214-030	M310B	4.9574	< 1.95E+00	1.18E-02	< 9.11E-01	< 5.88E-01	5.59E-04	9.03E-03	< 6.66E-02	< 1.0E+00	< 5.86E+00	< 2.14E+01	< 3.71E-01	
000214-031	M32	5.3215	< 1.81E+00	4.57E-05	< 8.48E-01	< 5.48E-01	7.59E-04	1.04E-02	< 6.20E-02	< 1.0E+00	< 5.46E+00	< 2.00E+01	< 3.45E-01	
000214-032	M58	5.0003	< 1.93E+00	9.10E-05	< 9.03E-01	< 5.83E-01	7.14E-04	1.23E-02	< 6.60E-02	< 1.0E+00	< 5.81E+00	< 2.13E+01	< 3.68E-01	
000214-033	M7	5.2248	< 1.85E+00	6.74E-05	< 8.64E-01	< 5.58E-01	2.10E-04	3.10E-03	< 6.32E-02	< 1.0E+00	< 5.56E+00	< 2.03E+01	< 3.52E-01	
000218-017	G158	5.4001	< 1.79E+00	7.53E-05	< 8.36E-01	< 5.40E-01	1.15E-02	2.20E-01	< 6.11E-02	< 1.0E+00	< 6.60E+00	< 1.97E+01	< 3.40E-01	1.46E+04
000218-018	G162	5.0720	< 1.90E+00	3.99E-05	< 8.90E-01	< 5.75E-01	1.20E-02	2.50E-01	< 6.51E-02	< 1.0E+00	< 5.73E+00	< 2.10E+01	< 3.63E-01	1.47E+04
000218-019	G181	5.0487	< 1.91E+00	3.00E-05	< 8.95E-01	< 5.78E-01	5.80E-03	1.18E-01	< 6.54E-02	< 1.0E+00	< 5.76E+00	< 2.11E+01	< 3.64E-01	2.00E+04
000218-020	G188	5.0793	< 1.90E+00	1.05E-04	< 8.89E-01	< 5.74E-01	1.31E-02	1.64E-01	< 6.50E-02	< 1.0E+00	< 5.72E+00	< 2.09E+01	< 3.62E-01	1.89E+04
000218-021	P1061	5.5693	< 1.73E+00	1.26E-04	< 8.11E-01	< 5.24E-01	1.54E-02	5.37E-01	1.63E-01	< 1.0E+00	< 5.22E+00	< 1.91E+01	< 3.30E-01	2.35E+04
000218-022	P1068	5.5736	< 1.73E+00	8.23E-05	< 8.10E-01	< 5.23E-01	1.08E-02	3.23E-01	1.63E-01	< 1.0E+00	< 5.21E+00	< 1.91E+01	< 3.30E-01	2.08E+04

Attachment 2.5
Analytical Results for Aluminum Button Samples (TBD)

ATTACHMENT 3

Script for Talking with Vendors who acquired gold from the Paducah Gold recycling program.

FIRST TALK TO RECEPTIONIST AND GET CONNECTED WITH A MANAGER OWNER ETC. THEN...

HELLO MY NAME IS TONY ACUNA WITH THE DEPARTMENT OF ENERGY. WE ARE IN THE PROCESS OF RESEARCHING METALS RECYCLING CONDUCTED AT THE DEPARTMENT'S PADUCAH PLANT. THIS RECYCLING WOULD HAVE INVOLVED SEVERAL METALS SUCH AS GOLD AND SILVER THAT WERE SOLD TO PROCESSORS SUCH AS YOU.

THESE SALES OF SURPLUS GOLD WOULD HAVE BEEN CONDUCTED VIA THE BID PROCESS BY UNION CARBIDE COMPANY AND LATER BY MARTIN MARIETTA ENERGY SYSTEMS. OUR SHIPPING RECORDS INDICATE THAT YOUR COMPANY RECEIVED GOLD FROM THE PADUCAH KENTUCKY PLANT. WE ARE INTERESTED IN LOCATING HISTORICAL RECORDS OF TRANSACTIONS FROM RECEIVING TO ULTIMATE DISPOSAL OF GOLD FROM YOUR ACTIVITY. CAN YOU HELP?
(WAIT FOR ANSWER)

INFORMATION/QUESTIONS:

DO ANY INVENTORY RECORDS EXIST OF THE RECEIPT OF GOLD FROM PADUCAH?
→ WAS ANY ANALYSIS OR TESTS OF THE GOLD BARS PERFORMED?
IF SO, IS THERE ANY DOCUMENTATION OF THE RESULTS OF THESE TESTS?
WAS THE GOLD REFINED OR REMELTED?
IF GOLD WAS REFINED, WHAT PROCESS WAS USED?
DO ANY RECORDS EXIST ON THE SALE OR USE OF THIS GOLD?
COULD YOU TELL US IN GENERAL TERMS HOW YOU USED THE GOLD, I.E., SOLD IT TO OTHERS OR USED IT INTERNALLY?
(HOW?)

DO ANY INVENTORY RECORDS EXIST OR DO YOU CURRENTLY HAVE ANY OF THIS GOLD ON HAND OR SOMETHING YOU MADE FROM IT?

POSSIBLE QUESTION AND PROPOSED ANSWER:

WHY ARE YOU DOING THIS RESEARCH? BECAUSE OF SEVERAL ALLEGATIONS BY FORMER EMPLOYEE'S CONCERNING PAST PRACTICES AND ENVIRONMENT, SAFETY AND HEALTH ISSUES STEMMING FROM THE COLD WAR ACTIVITIES AT THE PADUCAH SITE.

ATTACHMENT 4

POTENTIAL RADIATION DOSES ASSOCIATED WITH RECYCLED GOLD: INITIAL EVALUATION SUMMARY

1 Introduction

This document summarizes preliminary evaluations of the potential radiological health impact of contaminants that may have been present in recycled gold processed by the Paducah Gaseous Diffusion Plant (PGDP) Metals Recycling Program. Reasonable upper limit concentrations based on analyses of process residues and other materials were assumed for a number of exposure scenarios, and consequent radiation doses were calculated by standard methods.

1.1 General Assumptions

A number of assumptions were made for the sake of conservatism, that is, in order to yield dose projections that are very unlikely to have been exceeded in reality. As noted in Section 3 below, the higher of a number of surrogate measurements was assumed to represent contamination levels in the recycled gold. It was further assumed that for any application, recycled gold from PGDP was used directly, without any admixture of other gold present in the stream of commerce. It was also assumed that there was no subsequent refining or processing by end users or intermediaries prior to the use of the recycled gold.

2 Scenario Descriptions

2.1 External Dose (Jewelry)

As noted in Section 5 below, appreciable contamination of gold jewelry with naturally occurring radioactive materials (long-lived radon progeny) occurred well before the advent of modern nuclear technology. Skin damage was caused by prolonged contact with gold containing the high-energy beta emitter ^{210}Bi supported by its long-lived parent, ^{210}Pb .

A similar scenario was assumed for the present case. A large (50 cm^2) item of jewelry composed of 18 karat gold (75% gold) was assumed to have been worn full time over the same area of skin. The gold was assumed to contain the radionuclide concentrations described in Section 3, and beta doses to the underlying skin were assessed with the *VARSKIN* code at the standard depth of 0.07 millimeters (7 mg/cm^2).

2.2 Internal Dose (Injected Antiarthritic Compounds)

Of the several commercial applications considered, the use of gold as an injected antiarthritic agent produced the highest internal dose potential. Uranium and transuranic radionuclides are not readily absorbed through the gut, a factor that reduces the radiotoxicity of these materials when they are ingested. However, injection as a soluble compound bypasses the gut, and it is assumed that all of the injected material is taken up by the blood stream.

A number of injected gold compounds have been used to treat rheumatoid arthritis since the 1930's, among them gold thiomalate and gold thioglucose. Oral gold salts (e.g., auranofin) were introduced in the 1980s. The exposure scenario considered here was a standard course of treatment with gold thiomalate, consisting of a series of injections of increasing dose until a total dose of one gram was reached in the course of a year. The gold portion of the compound (approximately 50% by weight) was assumed to consist of recycled PGDP gold at the activity levels described below, with no dilution by other sources of gold, and no reduction in contamination during the process of formulating the drug.

Internal doses were assessed by applying the 50 year committed effective dose conversion factors from ICRP Publication 72 to the product of the mass of gold injected (0.5 g) and the concentrations of radionuclides presented in Table 3.1. The ingestion dose conversion factors presented in Table A.1 of ICRP Publication 72 were divided by the gut uptake factor (f_I) to obtain factors appropriate for the injection scenario.

3 Assumed Contamination Levels

The concentration of radioactive materials assumed in the following scenarios are based on the results of tests performed on various samples and solid residues.¹ A preliminary evaluation of potential radiation doses indicated that the results were sensitive to certain concentrations reported as "less than" values in the initial analysis. More sensitive reanalyses of archived samples yielded substantially lower "less than" values, and in some cases, positive results. Table 3.1 summarizes the concentrations assumed for the dose evaluation scenarios. Also presented (in small italics) are substantially lower concentrations that are more likely to reflect actual concentrations in the recycled gold.

¹ "Summary of Radiochemical Data for Paducah Gold Samples" Draft report by USDOE, 2000.

However, for the purpose of presenting a reasonable "worst case" analysis, dose projections were based on the higher concentrations noted in the combined leachates and digestions of the indicated process residues (crucible solids).

Table 3.1

Radionuclide	Concentration	Units	Material
U_{total}^*	1,496	pCi/g	Black Crucible solids 69977
^{99}Tc	32,000	pCi/g	Black Crucible solids 69968
^{99}Tc	<446	pCi/g	Refined Gold from process sample
^{237}Np	34**	pCi/g	Black Crucible solids 69977
^{237}Np	0.11	pCi/g	Process sample (Gamma spectroscopy)

* Ratios of ^{235}U to ^{238}U indicate that uranium was at or below natural enrichment. Natural isotope abundance was assumed for conservatism in dose calculations.

** This is the sum of a positive detection in the leachate (23.6 pCi/g) and the detection limit for the aqua regia digestion (10.4 pCi/g).

4 Results

4.1 External Dose

The *VARSKIN* calculation for the 50 cm² item of jewelry yielded a dose rate of 0.8 millirads per hour to the cells 0.07 millimeters directly under the jewelry. The resulting annual effective dose is 0.22 millirem per year. The effective dose takes into account the 1% weighting factor for the skin recommended in ICRP Publication 60 and the area averaging for stochastic effects recommended in ICRP Publication 59.

The local dose to the skin at 0.07 millimeters (shallow dose) is 7.2 rads per year, considerably below the ICRP Publication 60 recommendation of 50 rads per year to limit local skin damage.

4.2 Internal Dose

The committed effective dose projected for a one gram course of therapy with gold thiomalate is approximately 30 millirem, with approximately equal contributions from the uranium and neptunium concentrations shown in Table 3.1. This dose estimate is based on the assumption that the entire amount administered over a year was compounded from recycled gold. A dose of 30 millirem per year is approximately 10% of the effective dose from the natural background.

5 Historical Context

In the past, gold contaminated by medical isotopes has been used in the manufacture of jewelry, and some localized skin damage has been documented as a result. This problem predates the modern nuclear era, with the earliest likely case dating back to 1910.² Thin gold tubes loaded with several millicuries of ^{222}Rn (*gold seeds*) were sometimes used as substitutes for ^{226}Ra needles in radiotherapy and other applications. The gamma radiation produced was the same quality as that of a radium needle, but the 3.8 day half-life of radon made the gold seed more convenient for some procedures, including permanent implants.³

Unused or recovered gold seeds lost essentially all of their penetrating gamma radiation within a month of manufacture. The buildup of ^{210}Pb , a radon descendent with a half-life of 21 years, was sometimes not recognized, and some of the spent gold seeds were recycled, some into jewelry. In the 1960s, a number of reports of injuries due to such jewelry appeared in the medical literature.⁴ A campaign conducted by the New York State Department of Health in 1981 located over 150 pieces of contaminated jewelry and other objects out of about 160,000 items surveyed.⁵ Most of the contaminated objects dated from the 1930's or 1940's, although one gold ring was dated to 1910.

5.1 External Dose Rate from Jewelry Contaminated by Medical Gold Seeds

The *VARSKIN* calculations described above were repeated for the case of a gold ring contaminated with ^{210}Pb and progeny by the recycling of spent gold seeds. The beta radiation from ^{210}Bi is considerably more penetrating than that of the ^{99}Tc considered above, and the estimated concentrations were much higher. (Simon and Harley⁴ reported

² "Unwanted radioactive sources in the Public Domain: A Historical Perspective", J. O. Lubenau, *Health Physics* 76 (Operational Radiation Safety Supplement) pp. S16 – S22, February, 1999.

³ "Radon: Its Technique and Use", W. A. Jennings and S. Russ, Wyman & Sons, Ltd, London, 1948.

⁴ e.g., "Skin Reactions from Gold Jewelry Contaminated with Radon Deposit", N. Simon and J. Harley, *JAMA* 200:254-255, 1967.

⁵ "Report to the Governor and Legislature – Radioactive Gold Jewelry". New York State Department of Health, September, 1972

that as many as 100 spent gold seeds may have been used in a ring. The residual ^{210}Pb activity of each seed would have been about 0.5 microcuries, based on the typical 2 mCi ^{222}Rn loadings described by Jennings and Russ.³

The *VARSKIN* code yielded a shallow skin dose rate in excess of 1,000 rads per year for this case, considerably greater than the ICRP recommendation of 50 rads per year, and consistent with the types of injury reported in the literature.

5.2 Potential Internal Doses from Gold Contaminated by Radon Daughters

The use of gold injections to relieve arthritic symptoms dates back to the 1930s, and coexisted with the use of radioactive gold seeds for several decades. However, the relatively small quantities used in each application, as well as the refining steps involved in drug manufacture, diminishes the likelihood that full-strength spent gold seeds were compounded directly into a drug. At any rate, no account of the incorporation of spent gold seeds into antiarthritic compounds has been located.

For context, however, the same worst case internal dose scenario considered above for DoE gold was applied to spent gold seeds. The resulting dose estimate for the injection of 1 gram of gold thiomalate compounded of spent gold seeds is in excess of 4,000 millirem, more than 100 times the hypothetical dose calculated for recycled PGDP gold.

6 Conclusions

A reasonable worst-case analysis of potential radiation doses associated with recycled PGDP gold indicates that any such doses would have been small. It is quite unlikely that effective doses in excess of about a tenth of the annual natural background would have occurred had the recycled gold been used directly in commerce, without admixture or refining. These conclusions are largely based on the analysis of process residues, and are subject to revision if some of the actual product can be located and analyzed.

ATTACHMENT 5

	PGDP - Product Recycled and Sold				
	<u>Aluminum</u>	<u>Nickel</u>	<u>Shredded Lead</u>	<u>Monel</u>	<u>Cobalt *</u>
"Sold" - No Date	2,325,790	2,261,340		781,540	20,800
1970	113,784				
1971	348,896				
1972	172,524				
1973	223,806				
1974					
1975	539,573				
1976	247,676				
1977		884,461			
1978	268,935	4,302,631			
1979		5,079,955			
1980	169,491	2,986,375			
1981	88,496	1,181,495	258,990		
1982	1,184	216,518			
1983		63,585			
1984					
1985					
1986	40,817			100,900	
1991		1,820			
TOTALS:	4,540,972	16,978,180	258,990	882,440	

* 50,700 pounds were indicated "shipped to Oak Ridge"